ISOTOPES AND RADIATION TECHNOLOGY

A Quarterly Technical Progress Review

Prepared for DIVISION OF TECHNICAL INFORMATION / DIVISION OF ISOTOPES DEVELOPMENT, UNITED STATES ATOMIC ENERGY COMMISSION, by P. S. BAKER, A. F. RUPP, and ASSOCIATES, OAK RIDGE NATIONAL LABORATORY

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TECHNICAL PROGRESS REVIEWS

To meet the needs of industry for concise summaries of current atomic developments, the Atomic Energy Commission is publishing this series, Technical Progress Reviews. Issued quarterly, each of the reviews digests and evaluates the latest findings in a specific area of nuclear technology and science.

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Isotopes and Radiation Technology

is a Technical Progress Review prepared by the Isotopes Information Center, Oak Ridge National Laboratory, at the request of the Divisions of Technical Information and Isotopes Development, U. S. Atomic Energy Commission. This Review is intended to assist those interested in keeping abreast of significant developments in the fields of isotopes and radiation technology. It is not a comprehensive review of all literature published in this field during a given quarter; rather, it is a mechanism for presenting concise, selected reviews of information on subjects of prevailing Commission interest as it becomes available.

This Review attempts to relate the published results of research and development sponsored by the Division of Isotopes Development to significant developments in radioisotopes and radiation technology, as reported in the world literature. Coverage includes isotope production and development, isotope technology development, isotope applications technology, process radiation development, and radiation processing of food and medical supplies.

In addition to reviews of current literature and special review articles dealing with specific isotopes, facilities, and applications, this publication occasionally contains feature articles prepared by recognized experts on specific topics of current interest. Critical evaluations and interpretations presented are those of the editors and invited reviewers; therefore readers are encouraged to consult the original references in order to obtain all the background of the work reported and the interpretation of the results given by the original authors.

The subject headings listed below have been adopted to help maintain continuity and order, from one issue to the next, in the material reviewed. All reviews, except Feature Articles, will be arranged under these headings; but any one issue will not necessarily contain all the headings.

Isotope Production and Development
Isotope Technology Development
Food Irradiation
Process Radiation Development
Source Development
Isotopic Power Development
Applications in Life Sciences
Safety
Isotopes Around the World
Miscellaneous

P. S. Baker, Robert H. Lafferty, Jr., Martha Gerrard,
R. E. Greene, F. E. McKinney, and Joan E. Carden, *Editors*
A. F. Rupp, *Adviser*

Oak Ridge National Laboratory

Section

I

Isotopes and Radiation Technology

Isotope Production and Development

Isotope Production at General Electric's Vallecitos Nuclear Center

By N. C. Howard*

Abstract: The radioisotope production facilities of General Electric's Vallecitos Nuclear Center are described. Details of the General Electric Test Reactor presented include neutron-flux data, isotope production positions in the core, hydraulic shuttles, and cable devices for irradiation alongside the core. Postirradiation procedures and a summary of currently produced radioisotopes are also given.

*General Electric Company, Vallecitos Nuclear Center, Pleasanton, Calif.

Completed in late 1958, the General Electric Company's Vallecitos Nuclear Center (Fig. 1), located near Pleasanton, Calif., has played a major role in furthering the peaceful uses of atomic energy. Since 1959 the Irradiation Processing Operation has been producing and marketing many high-specific-activity radioisotopes for research and development, industrial radiography, medical therapy, and radiopharmaceutical and other industrial and medical applications.

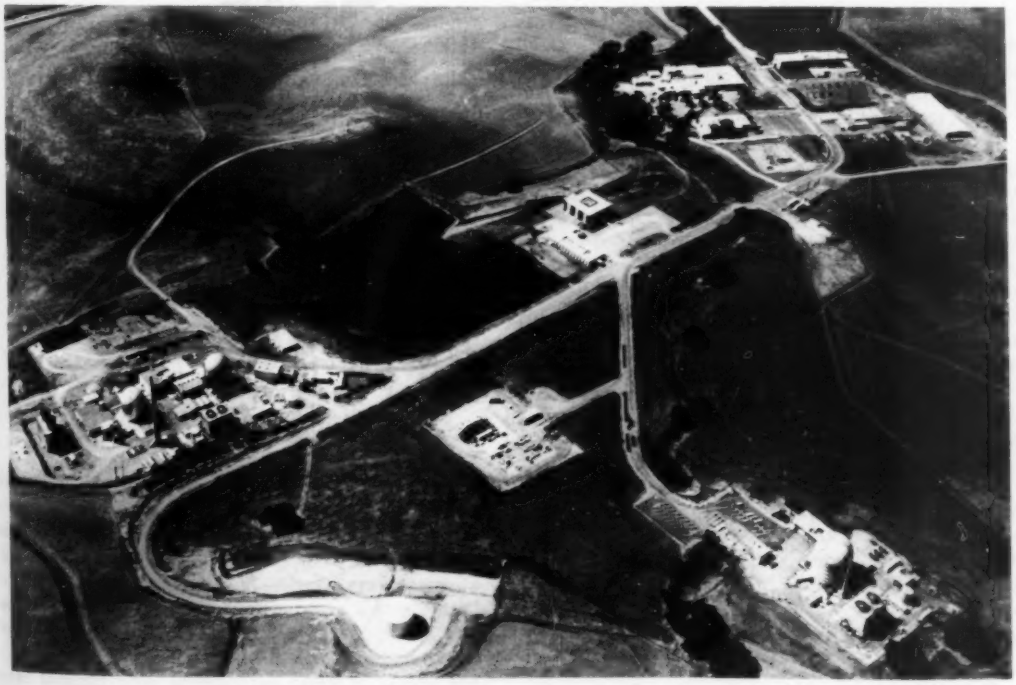

Fig. 1 Vallecitos Nuclear Center, Pleasanton, Calif.

Facilities available at the laboratory include a 50-Mw General Electric Test Reactor (GETR), a 30-kw Nuclear Test Reactor (NTR), a Radioactive Materials Laboratory, a Nuclear Chemistry Laboratory, a Nuclear Physics Laboratory, a Thermionic Conversion Laboratory, a Plutonium Fuel Fabrication Facility, an Isotope Production Laboratory, a Nuclepore Filter Development Laboratory, and a capsule fabrication and assembly shop.

Reactor Facilities

GENERAL ELECTRIC TEST REACTOR

The GETR, which began operation in February 1959 as the first commercial test reactor, consists of a high-flux tank type pressurized-water reactor, a reflector pool, a storage and service canal, and other support and experimental facilities (Fig. 2). The unusual high-flux characteristics and operating schedule

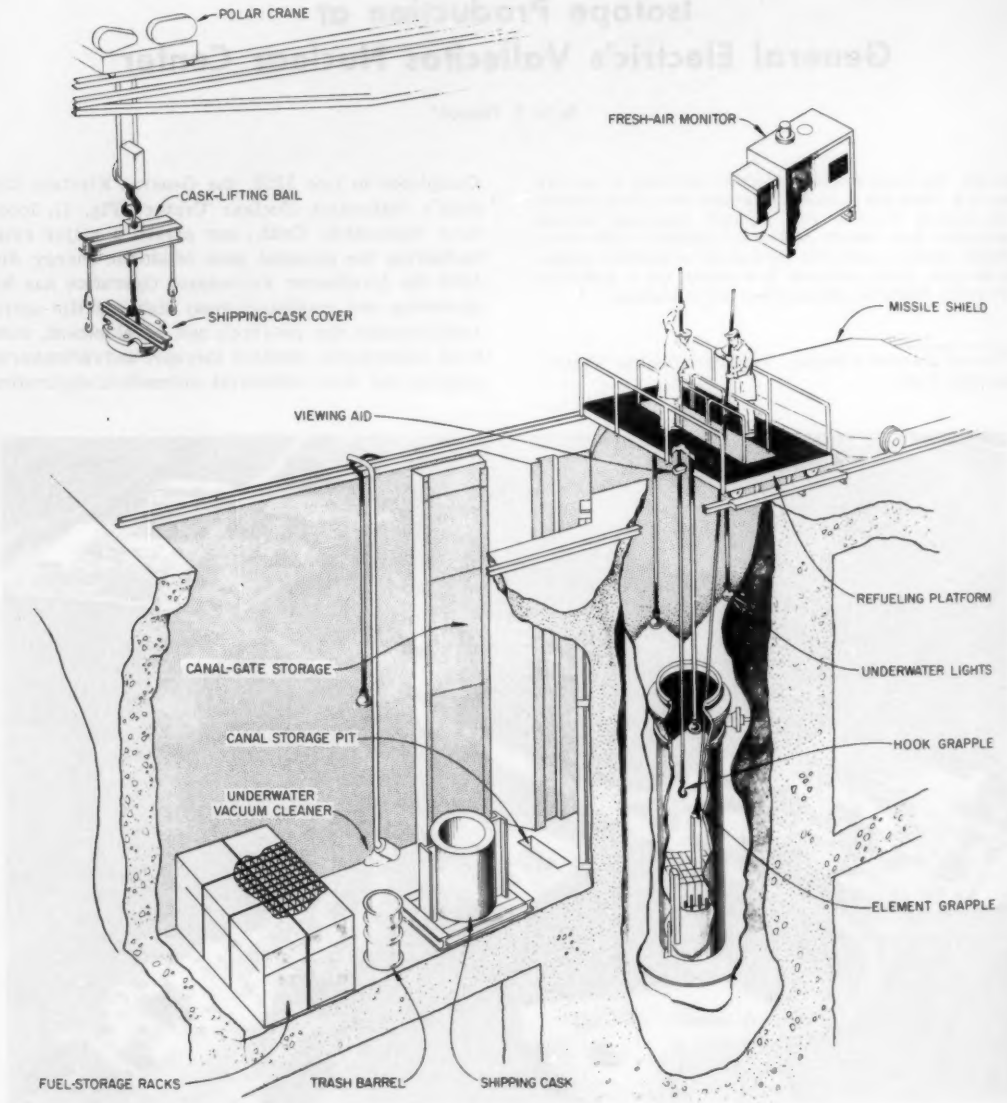

Fig. 2 The GETR canal and reactor-pool arrangement.

of the GETR permit the preparation of high-quality isotopes, some with specific activities not previously available in a commercial reactor. The reactor data of interest for the production of isotopes are:

Operating power	50 Mw (raised from 33 Mw in October 1966)
Thermal-neutron flux	Average 2.5×10^{14} n/(cm^2)(sec) Maximum 6.0×10^{14} n/(cm^2)(sec)
Fast-neutron flux (>0.18 Mev)	Average 1.8×10^{14} n/(cm^2)(sec) Maximum 4.0×10^{14} n/(cm^2)(sec)
Operating cycle	5 weeks: 28–31 days at full power and 4–7 days of shutdown for maintenance, experiment changes, and refueling
Midcycle refueling	One 10-hr shutdown every cycle
Operating efficiency	>70%

The reactor core is a pressurized matrix, moderated and cooled with light water, and has a 3-ft active length housed in a 2-ft-ID aluminum pressure vessel. The reactor operates with fuel, control rods, filler pieces, or through-core loops in the thirty-seven 3- by 3-in. core positions. Six control rods are positioned to operate permanently (Fig. 3). Fuel normally occupies twenty of the positions surrounding the control rods. Square beryllium or aluminum filler

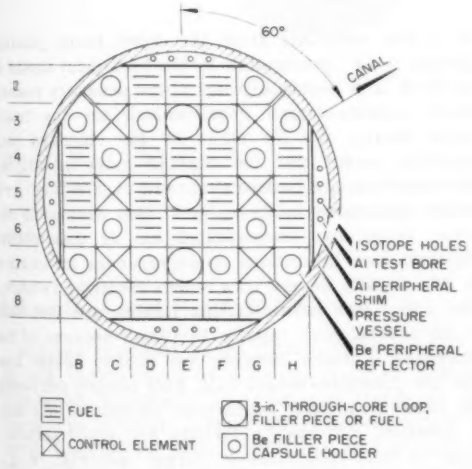

Fig. 3 The GETR core plan.

pieces occupy matrix positions not used by control rods, fuel elements, or through-core loops and are interchangeable with the reactor fuel elements. Curved aluminum and beryllium peripheral reflector pieces surround the fuel–filler array, provide neutron reflection, and shape the core into a cylinder.

A typical filler piece has a 1.5-in.-diameter hole bored longitudinally through the piece to accept the capsules or capsule baskets. Capsules can be installed in the core, directly in the filler pieces, or in a capsule basket in the filler piece. Special four-hole beryllium filler pieces are used in place of the standard filler pieces to increase the number of experimental locations in the core. One hole is the standard size, one is 1.25 in. in diameter, and the remaining two are 0.625 in. in diameter. Capsule baskets are particularly useful if several capsules are to be loaded in the same core filler piece.

The in-core positions available for isotope production account for approximately 7% of the total core volume. The unperturbed peak thermal-neutron flux in the core positions is 6.0×10^{14} n/(cm^2)(sec) with a fast-to-thermal neutron-flux ratio of 0.7. All experiments in the core are irradiated in a 4-week cycle or multiples thereof.

The pool, in which the core-containing pressure vessel is positioned, provides experiment space, gamma shielding, and emergency cooling water for the core. The pool facilities include fixed and adjustable capsule positions, one hydraulic shuttle, and one trail-cable tube* (Fig. 4). Since the pool has more free space than the core, capsules of various sizes and shapes can be irradiated in the pool. There are 42 capsule tubes 1.5 in. in diameter and 45 in. long outside the reactor pressure vessel. Larger tubes, each occupying two small-tube spaces, are also used. The average unperturbed thermal-neutron flux in the pool facility is 1.2×10^{14} n/(cm^2)(sec) with a peak flux of 2.2×10^{14} n/(cm^2)(sec) in certain locations. Irradiation time for these bulk pool irradiations is the same as for the experiments in the core. Isotopes produced in the pool positions are those requiring large quantities of target material, such as ^{60}Co, ^{14}C, and ^{131}I.

The hydraulic shuttle-tube facility (Fig. 5) is a device for rapidly positioning small capsules in, and removing them from, the neutron-flux zone adjacent to the reactor pressure vessel. Irradiations from a few seconds to several days can be achieved without interrupting reactor operation. The facility consists of a tube extending from the pool support system upward through the pool wall to the capsule catch basket in the canal. The inserted shuttles end in one of seven vertical positions that extend from the bottom to the top of the reactor core. This facility is used in the production of short-half-life high-specific-activity isotopes such as ^{99}Mo, ^{197}Hg, ^{198}Au, and ^{131}I.

A trail-cable facility (Fig. 6), located in pool capsule positions Z13 and Z14, is available for capsule irradiations. This facility, which, like the shuttle tube, allows relatively short irradiations without interruption of normal reactor operation, consists

*"Trail cable" is General Electric's descriptive name for a facility in which targets are trailed at the end of a cable — *The Editor*.

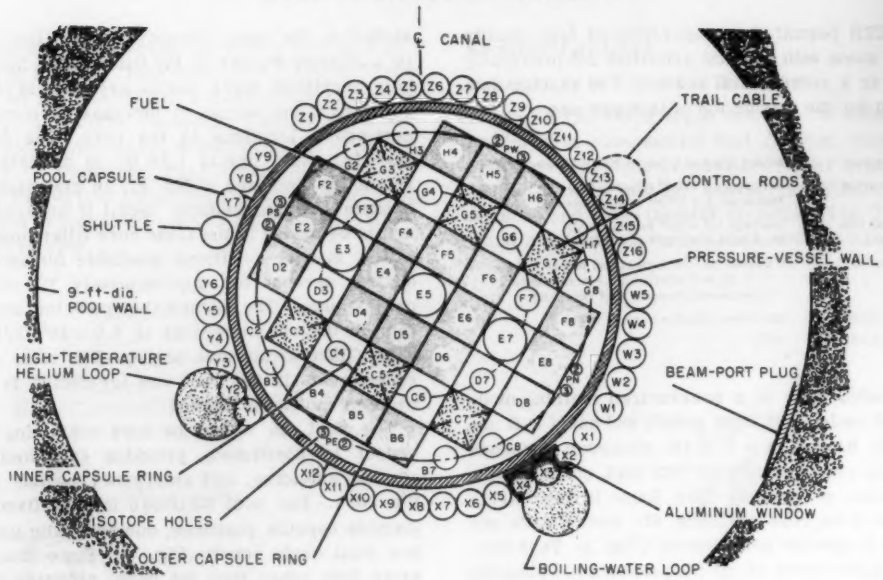

Fig. 4 Radial locations of GETR experiment facilities.

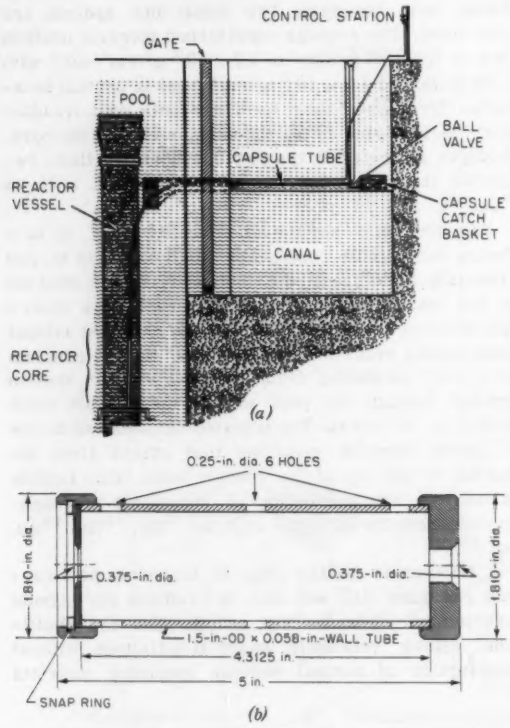

Fig. 5 (a) Hydraulic shuttle facility and (b) standard shuttle-tube carrier.

of a tube extending from the third floor, passing through a 45° penetration in the biological shield to the pool, and ending adjacent to the pressure vessel. Basic capsule-design requirements for the trail-cable facility are the same as for standard pool capsules except that the capsule must be adapted for insertion and withdrawal with a flexible trail cable. Variations in the vertical flux during the reactor cycle are compensated for as they occur. Physical size limitations on capsules for irradiation in this facility depend on a length/diameter relation. The actual flux values in this facility are less than in the normal pool capsule facilities because of the increased distance between the facility center line and the pressure-vessel wall. This facility performs the same irradiation function as the shuttle tube and, in addition, provides a facility for short irradiations of bulk quantities of target material, e. g., $^{124}Xe(n,\gamma)^{125}Xe \xrightarrow{EC} {}^{125}I$.

A new shuttle-tube facility for capsule irradiations in the core has recently been installed to permit production of high-specific-activity ^{99}Mo, ^{197}Hg, and other isotopes currently in demand by the radiopharmaceutical industry. This facility, which will permit insertion and removal of capsules from the reactor core without interrupting normal reactor operation, will have a shuttle tube 1.25 in. in diameter, and capsules will be loaded and discharged in the service and storage canal about 10 ft underwater. Capsules will be hydraulically forced down a guide tube into a receiver section in the pressure vessel near the top of the reactor core and then mechanically

lowered into the active core region at 4 in./min to prevent uncontrolled reactivity changes. The capsules will be discharged by raising them into the receiver section and reversing the water flow. The average unperturbed thermal-neutron flux available in the in-core shuttle will be 2.6×10^{14} n/(cm^2)(sec), and the average fast-neutron flux will be 1.8×10^{14} n/(cm^2)(sec).

of high-specific-activity isotopes. In addition to those listed in Table 1, any reactor-produced isotope can be made, e.g., 18-min ^{88}Rb. Recently a sample of this isotope was shipped by helicopter to the University of California with a total time lapse of 19.5 min from discharge of the capsule from the reactor until the sample was being counted on the Berkeley campus, some 40 miles away.

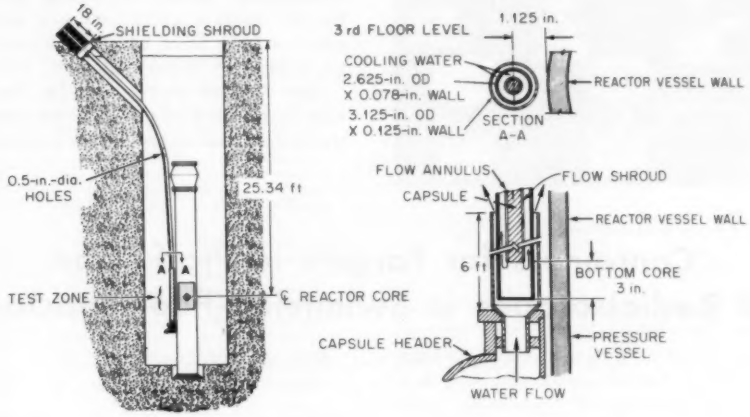

Fig. 6 The trail-cable facility.

THE NUCLEAR TEST REACTOR

The NTR is used only occasionally for isotope production; it is used most often for activation-analysis studies.

Laboratory and Hot-Cell Facilities

Isotopes produced in the GETR can be categorized as bulk, unprocessed, uncalibrated isotopes; bulk, unprocessed, calibrated isotopes; and processed isotopes. The isotopes in the first group are shipped directly from GETR, after irradiation, to customers off site. Isotopes in the other two groups are transferred to the Radioactive Materials Laboratory for calibration or processing or both. Isotopes to be calibrated, such as ^{60}Co, are prepared for shipment in one of four high-level cells. Processed isotopes are handled in the Isotope Production Laboratory. This laboratory also prepares high-purity natural and enriched targets for irradiation. Bulk and quartz targets (Fig. 7) are sealed in aluminum capsules for irradiation in the GETR. Postirradiation processing in the hot cell ranges from simple dissolution of the targets to complex chemical separations such as ion exchange and distillations. The isotopes listed in Table 1 are now being produced to provide a continuing inventory

Fig. 7 A Vallecitos technician sealing mercuric oxide inside a quartz ampule.

Table 1 PROCESSED ISOTOPES CURRENTLY PRODUCED AT THE VALLECITOS NUCLEAR CENTER

Isotope	Half-life	Specific activity, mc/g	Chemical form
^{32}P	14.7 days	Carrier free	H_3PO_4 in 1*M* HCl
^{45}Ca	165 days	3000–8000	$CaCl_2$ in 1*M* HCl
^{51}Cr	27.8 days	200,000–1,500,000	$CrCl_3$ in 1*M* HCl
^{59}Fe	45 days	15,000–45,000	$FeCl_3$ in 1*M* HCl
^{75}Se	120 days	150,000–900,000	H_2SeO_3 in 1*M* HCl
^{85}Sr	64 days	12,000–30,000	$Sr(NO_3)_2$ in 1*M* HNO_3
^{99}Mo	66 hr	>600	$(NH_4)_2MoO_4$ in conc. NH_4OH
^{131}I	8.05 days	Carrier free	NaI in basic sodium sulfite
^{197}Hg	65 hr	>3,000,000	$Hg(NO_3)_2$ in 1*M* HNO_3
^{203}Hg	47 days	8000–30,000	$Hg(NO_3)_2$ in 1*M* HNO_3

Isotope production in the GETR before 1966 was geared toward production of high-specific-activity long-half-life isotopes (i.e., greater than 7 days) in the core. Growing interest in several of the short-half-life isotopes by the radiopharmaceutical industry has stimulated efforts toward developing facilities for producing them, e.g., ^{197}Hg and ^{99}Mo. The production, processing, and distribution of these two isotopes to customers in the Midwest and on the East Coast have been made possible through the use of the shuttle-tube and trail-cable facilities, together with closely timed scheduling of shipments by air freight. Weekly shipments are made with a maximum of 30 hr from time of discharge from the reactor to delivery to the customer.

With the new in-core shuttle, the reactor will be able to produce isotopes having higher specific activities than those previously produced. (FEM)

Containers for Targets in Production of Radioisotopes in Swimming-Pool Reactors

By H. W. Harvey, J. C. Hoh, W. C. Mohr, and D. C. Stewart*

Abstract: One inexpensive can, of three types tested, was found to be possibly suitable for use as a target container in the production of radioisotopes in a swimming-pool reactor.

The THOR reactor at the Institute of Nuclear Science of the National Tsingua University in Taiwan is an open-pool light-water-moderated reactor with the core 25 ft below the water surface. Primarily owing to an inability to develop leakproof target containers, the institute staff was having difficulty in using the irradiation baskets adjacent to the reactor core for routine production of radioisotopes. At their request we investigated several containers, using the simplest and least expensive equipment possible in the hope that it would be usable by other reactor groups operating on limited budgets.

Materials and Equipment

Commercially available aluminum cans were studied on the assumption that their unit cost would be low enough to allow their use on a throw-away basis: a 211 × 413 beer can (Reynolds) and a 307 × 113 tuna and a 208 × 208 potted-meat can (Continental Can). Since all these cans are formed by the impact extrusion process, the only seam on the closed container is that formed in sealing on the lid. In our tests the seals were made with a home canner, Dixie SD-H-23 Hand Sealer, designed to seal one size can only (Dixie Canner Equipment Co., Athens, Ga.; current price $230). The cans were opened with a Sears-Roebuck Automatic Electric Can Opener (catalog 34A8345, $7.88). With slight modifications, i.e., extension of the operating handle and mounting on a heavy base, the opener can readily be operated remotely.

Procedure for Use

When in use for radioisotope production, the cans would be weighted, so that they would sink, loaded with target material, sealed, and placed in the irradiation baskets. After irradiation, the cans would be transferred to an underwater shelf on the side of the reactor pool, where they would be loaded into a shielded transfer cask having a water drain. The cask would be lifted from the pool, drained, moved to a shielded cell in the radioisotopes processing building, and opened. The cans would then be removed and opened with a kitchen type electric can opener and disposed of as active waste after the contents had been removed.

Tests on Cans

The results of pressure tests on unirradiated cans showed the potted-meat cans to be the most satisfactory of the three (Table 1, Fig. 1). The pressure tests were made by submerging the cans, containing 20 ml

*Argonne National Laboratory, Argonne, Ill. Sample cans for the tests were obtained through the kind efforts of John J. Daly of Continental Can Co., Inc., Chicago, Ill., and Roger P. Cornick of Reynolds Metals Company, Richmond, Va.

of water, in a saline solution under 10 to 40 psi pressure for varying times and then, by a silver nitrate or conductivity test, examining the water in the cans for salt that may have leaked in.

Although the unprotected beer cans crushed rather readily, they stood up well when a counterpressure was established inside the can by adding 1 g of Dry Ice before sealing (cans 3 and 4).

Meat cans that had received 7×10^6 rads of gamma radiation from a ^{60}Co source showed no leaks at pressures up to 20 psi. Since deterioration of the plastic coating, which contained chlorine, on the inside of the cans made use of the silver nitrate or the conductivity leak test impossible in this series, the sealed cans were submerged in kerosene in a desiccator, and leaks were considered absent if no bubbles formed when 28 in. Hg vacuum was drawn on the desiccator.

Meat cans sealed with a standard soft plastic gasket and irradiated in the Argonne CP-5 reactor for a week at a flux of 10^{12} n/(cm^2)(sec) also were free of leaks.

(MG)

Table 1 PRESSURE TESTS ON NONIRRADIATED CANS

Can type	No.	Pressure, psi	Time under pressure, min	Leak test		Condition of can
				$AgNO_3$	Conductivity	
Beer, 211 × 413	1	10	6	Leak	Leak	Crushed
	2	15	6	Leak	Leak	Crushed
	3*	15	30	Neg.	Neg.	No change
	4*	20	2 × 60	Neg.	Neg.	No change
Tuna, 307 × 113	5	20	15	Neg.	Neg.	No change
	6	25	35	Neg.	Neg.	Crushed
	7	30	31	Neg.	Neg.	Crushed
	8	25	60	Neg.	Neg.	Crushed
	9	25	60	Neg.	Neg.	Crushed
	10	25	60	Neg.	Neg.	Crushed
	11	20	89 × 60	Neg.	Neg.	No change
	12	20	89 × 60	Neg.	Neg.	No change
	13	20	89 × 60	Neg.	Neg.	No change
	14	40	20	Neg.	Neg.	Crushed
Potted meat, 208 × 208	15	30	2	–	–	Crushed
	16	20	45	Neg.	Neg.	No change
	17	25	60	Neg.	Neg.	No change
	18	20	65.5 × 60	Neg.	Neg.	No change
	19	20	65.5 × 60	Neg.	Neg.	No change
	21	20	6.5 × 60	Neg.	Neg.	No change
	22	20	6.5 × 60	Neg.	Neg.	No change
	23	20	6.5 × 60	Neg.	Neg.	No change

*Approximately 15 psi pressure inside can from CO_2 gas evolved from Dry Ice.

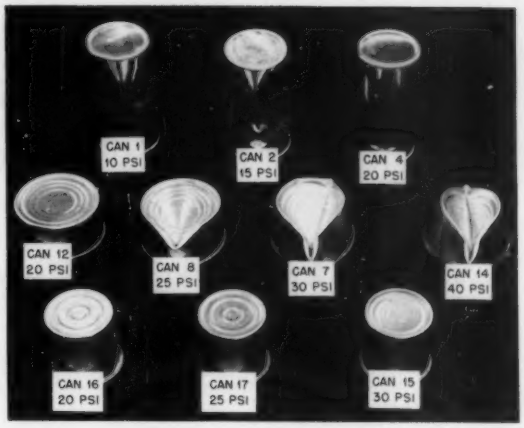

Fig. 1 Nonirradiated cans after pressure tests.

Section

II

Isotopes and Radiation Technology

Isotope Technology Development

Prototype Radioisotope Inspection System for Measurement of Process Residues on Electronic Circuit Boards*

By I. Sturman and I. J. Wright†

Abstract: Residual solder flux on automatically processed electronic circuit boards has been quantitatively and routinely determined by adding a radioisotope tracer to the flux and counting the board after processing. Less than 1% of the flux remained after processing to the applicable specification. The automatic solder process, the large-area plastic scintillator beta detector that was designed and fabricated for the measurements, and the calibration techniques are described. Health and safety techniques and training procedures required because the study was performed in a production area with production personnel were shown to be economically reasonable; thus the techniques of radioisotope tracing may be considered a production-control tool.

Tracerlab/Richmond, under a contract with the USAEC Division of Isotopes Development, has designed, developed, tested, and installed a prototype radioisotope inspection system to routinely and quantitatively detect and determine the amount of solder-flux residue on electronic circuit boards in an effort to improve their reliability. The proposed method was evaluated under production-line conditions, with production-line personnel, under a cooperative arrangement between Tracerlab/Richmond and the Autonetics Division of North American Aviation, Inc., at its facility in Anaheim, Calif. Reference 1 is a complete report of this study. The system was designed in accordance with principles developed under a previous contract.[2]

This article first briefly covers the circuit-board production process that was studied; next the important findings of the preceding study of the entire process are summarized; then the prototype routine radioisotope inspection system that was developed and its use are described; finally the personnel protection and training procedures required in the use of radioisotopes in production-control problems are discussed.

Circuit-Board Production Process

With the present emphasis in the aerospace industry on miniaturization and long-term high reliability, the manufacture of system elements for such complex weapon systems as the Minuteman missile is placing extreme demands on existing inspection and quality-control methods. Printed circuit boards, with hundreds of solder joints and precision-plated circuit lines, present a particularly difficult problem in control of reliability. Starting with simple terminal boards, the industry has evolved through single-sided printed circuit boards, high-density double-sided boards, and three-dimensional potted modules to multilayer boards in which as many as 20 thin circuit layers are laminated together in an integrated electronic package.

The total quantity of printed circuit boards of all types manufactured in 1962 has been estimated at over 100 million, with an aggregate value of over $\$20 \times 10^9$. The production of these printed circuit boards under high-reliability specifications results in the initial rejection of a large percentage, which must be reprocessed before acceptance.

A basic reason for this rejection rate is that process materials, such as soldering flux, "resist" lacquer, and ferric chloride etching solution, are present as residual low-level contaminants on the finished boards. This residual contamination is responsible for poor quality and reliability in the following ways: defective connections in solder joints, short-circuiting due to absorbed moisture and electrolytes, corrosion of the conductor lines, and corrosion of the solder joints along the solder-plating interface. These and other effects occur over an

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extended period, and, even though a board is passed by an existing inspection technique, it may fail in the field under operating conditions.

Each solder connection, each conductor line, and any given surface area can cause a defective board if contaminated by residual process materials. Because of the great complexity of joints and components, this contamination problem cannot be controlled with any high degree of confidence by techniques that rely primarily on visual methods. No matter what optical aids are provided, these techniques remain time consuming, fatiguing, and highly dependent on the subjective opinion of the observer.

Fluorescent penetrants and ultraviolet light have been used by other investigators[3] in a system for inspecting soldered electrical joints. They showed that their system extended the capability of visual inspection by revealing voids and defects not visible under normal lighting; however, the system was not completely successful because during destructive testing a substantial number of joints judged to be faulty because of surface faults had strengths greater than the acceptable level. The system required application of a penetrant on the cleaned boards, removal of excess penetrant with an emulsifier, application of a developer to draw out the penetrant, and viewing of the parts under ultraviolet light in a hood to exclude ambient light. The process and the machinery for application of the various chemicals were somewhat complicated for results that could only be qualitative and subject to operator interpretation.

The type of board chosen for the Tracerlab study was the double-sided plated-through-hole etched board. This was the most common board used in the aerospace industry at the time of the study. The gold-plated conductor system was chosen since it represented the system used in both the Minuteman and Polaris programs. Figure 1 represents schematically the general production process of such a board.

Preliminary Study

Tracerlab/Richmond made a preliminary study of applications where radioisotope tracers might be used profitably in the circuit-board production process. The specific purposes of this study were:

1. To investigate with radioactive tracers the effect of very-low-level process contamination introduced onto the surface of printed circuit boards and to correlate these levels of contamination with solder-joint quality.

2. To develop instrumentation and techniques for use of radioisotopes as routine quality and reliability inspection tools in circuit-board manufacture.

3. To develop adequate health physics procedures for using such tracers routinely with production personnel.

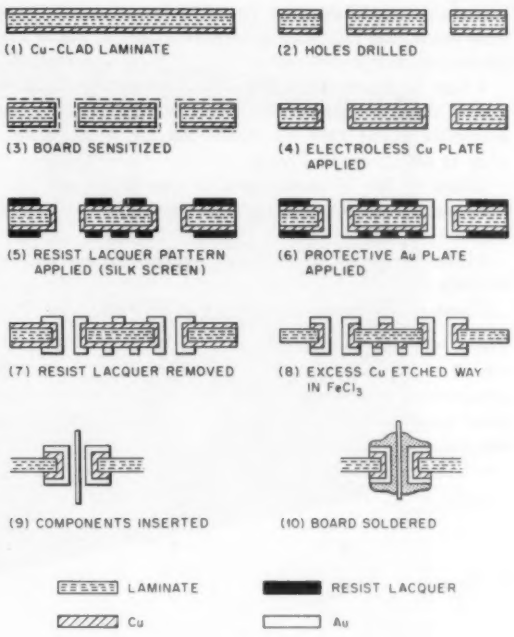

Fig. 1 Steps in production of plated-through-hole etched printed circuit board.

The final report[2] indicates that these goals were met successfully.

The three specific process materials studied as potential contaminants were:

1. The vinyl resist lacquer applied in step 5 (Fig. 1) to protect areas from gold plating.

2. The etching solution of $FeCl_3$ used to remove copper in step 8.

3. The solder flux, an aqueous solution of glutamic acid hydrochloride, applied prior to step 10.

These studies showed that ^{90}Y (half-life 64.4 hr, 2.18-Mev beta, no gamma) as eluted from the Brookhaven National Laboratory (BNL) ^{90}Y generator could be used as a satisfactory tracer for the first and third of these solutions, but it was of doubtful value as far as the $FeCl_3$ solution was concerned and ^{59}Fe was used instead. As a result of this study, several of the process specifications were changed to give better removal of process residues.

As part of this study, boards processed with tagged flux were counted and subjected to a humidity cycle test. [Specified as MIL-E-52720, this is a 240-hr test in which samples are cycled between room temperature and 71°C at a relative humidity of 95%. These conditions represented the most severe such specification in existence at that time (1963).] After the test, the boards were removed from the humidity

chambers and classified on the basis of visible corrosion into the following arbitrary classes, which are plotted against count rate in Fig. 2:

Class A: Heavy white deposits over entire surface; green deposits around conductors and holes. The minimum count rate for these boards, 2700 counts/min, is not shown in Fig. 2.

Class B: Heavy white deposits over entire surface.

Class C: Medium but uniform deposits over entire surface.

Class D: Light nonuniform deposits over entire surface.

Class E: No visible corrosion products.

These studies showed that scanning a board to localize the residual flux was not necessary but that integrating the counts over the entire board was quite valid and that the corrosion rate depended on the amount of flux on the board.

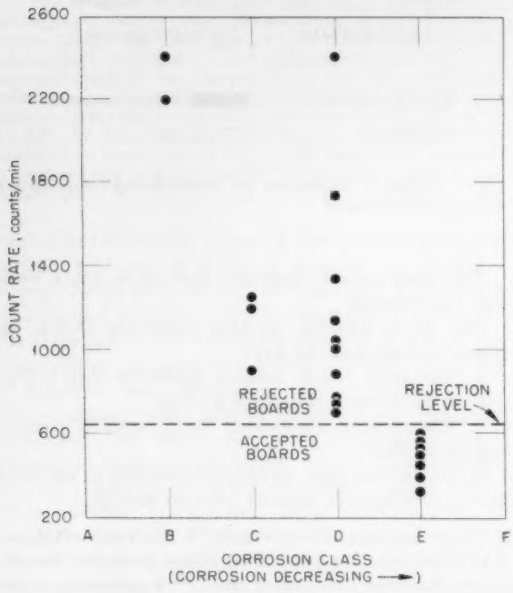

Fig. 2 Effect of residual flux (indicated by radioactivity) on corrosion of printed circuit boards after submission to MIL-E-52720 humidity test for 240 hr.

Prototype Inspection System

The results obtained in the preliminary study showed that radioisotope tracers would be an ideal tool for detecting traces of various residues in the manufacture of circuit boards. Therefore a decision was made to develop a system to test the tracer method on a routine production line. The detection of solder-flux residue was chosen because the relation between corrosion and residue had been so clearly demonstrated in the original test.

The Autonetics Automatic Dip Solder Machine[4] (Fig. 3*a*) was selected for this test. The function of this machine is to control precisely the deposition of solder onto etched circuit boards (Fig. 3*b*). The circuit boards are loaded onto carriers, which are moved along a track by a worm gear. The boards are dipped into the solder flux, allowed to drip, and then dried over electrical heaters. If the boards have excess moisture on them, voids can occur in the solder because of the steam generated by the heat of the molten solder. After drying, the boards are dipped into the solder and removed from the machine. The board carriers are reused. The boards are placed in a commercial dishwasher, where the flux is removed with detergent and deionized water. After a rinse with deionized water, the boards are dried either at room temperature or with heated air.

The above process was followed in conformance with Autonetics solder specifications. The flux, glutamic acid hydrochloride, has excellent soldering characteristics, and therefore its use is mandatory for high-reliability circuit boards. Unfortunately what makes for excellent soldering also results in a corrosive residue which must be completely removed.

DETECTOR SYSTEM

The heart of any routine radioisotope inspection method is the instrument used to detect the radioactivity. The previous study had shown that scanning the entire board was not necessary to localize the flux residue but integrating the counts over the entire board was quite valid. Since a detector was needed with as large an area as possible to process as large a circuit board as possible, a large-area plastic-phosphor beta detector (about 700 cm^2) was designed to be used with the system.

The primary advantage of a plastic-phosphor beta detector is that photomultiplier tubes and phosphors are commercially available in larger sizes than Geiger tubes. In this case the detector background from the unshielded 6-in.-diameter plastic phosphor was about 200 counts/min. This compares favorably with about 60 counts/min background from an unshielded 1-in.-diameter Geiger tube.

Another advantage of the plastic-phosphor beta detector over a Geiger counter is that the photomultiplier does not become saturated at high count rates. During the study the count rates observed on the boards immediately after removal from the automatic soldering machine averaged 142 thousand counts/min. Had Geiger tubes been used, this rate could not have been observed so accurately.

The original design area of the plastic phosphor was 6 by 6 in. An acrylic-plastic light pipe was designed to adapt the phosphor to a 5-in. photomultiplier tube, but, after the detector was fabricated, it

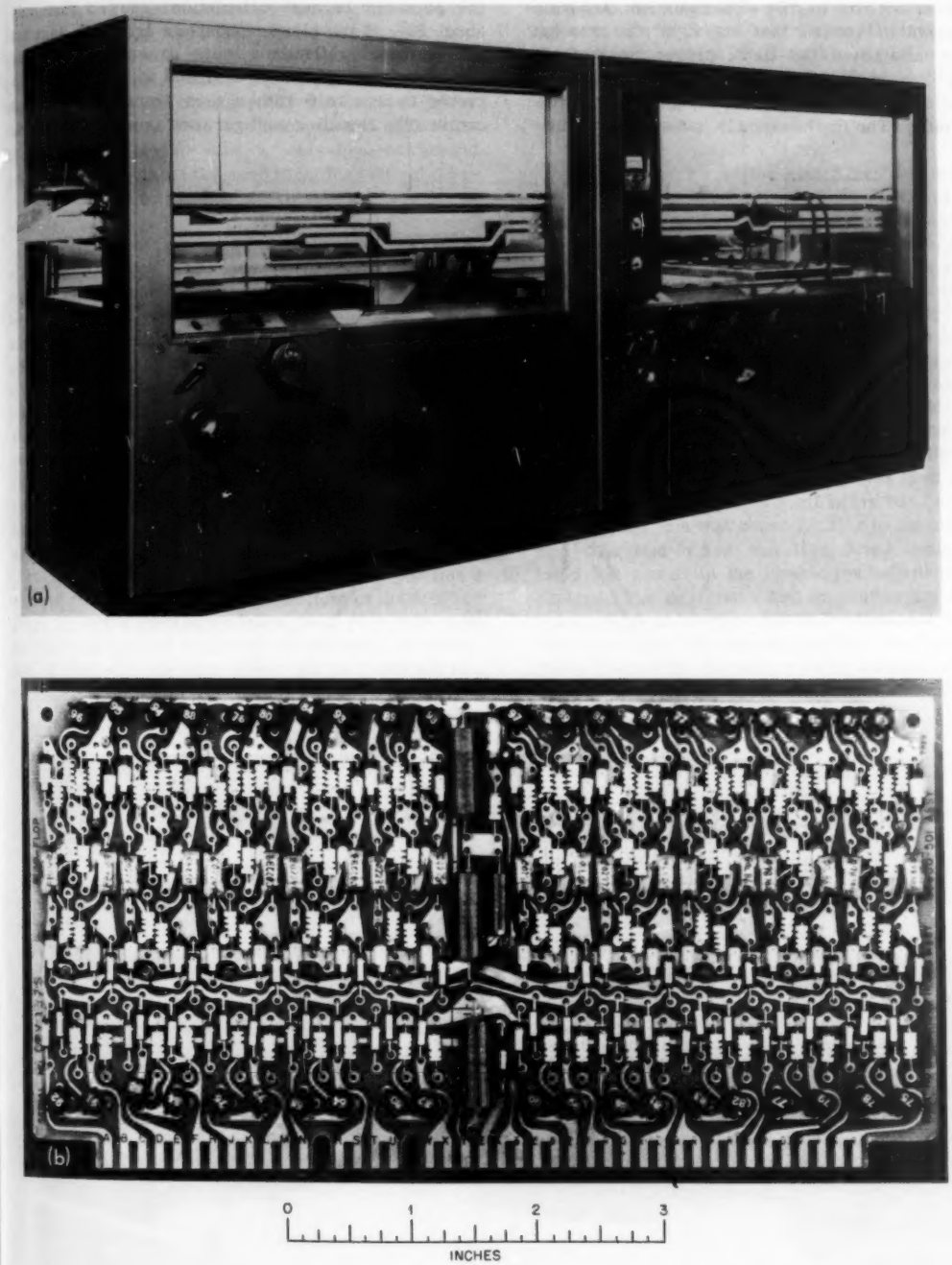

Fig. 3 (a) Autonetics automatic solder machine showing flux tank, drying section, and solder tank. Note that the machine is completely enclosed by glass or metal except at the ends, where boards are inserted or removed through small openings. (b) High-component-density circuit board, typical of boards processed in this machine.

had poor sensitivity to $^{90}Sr-^{90}Y$ radiation. Analysis of the problem showed that the light pipe was too long and attenuated the light, preventing it from reaching the photocathode. Therefore the plastic phosphor and light pipe were redesigned to improve the efficiency. The final assembly had a 6-in.-diameter phosphor because calculations showed that only about 20% of the plastic-phosphor area was lost but that internal reflections were drastically reduced when the light pipe was designed to reduce from a circle to a circle rather than from a square to a circle. The resulting configuration is shown in Fig. 4.

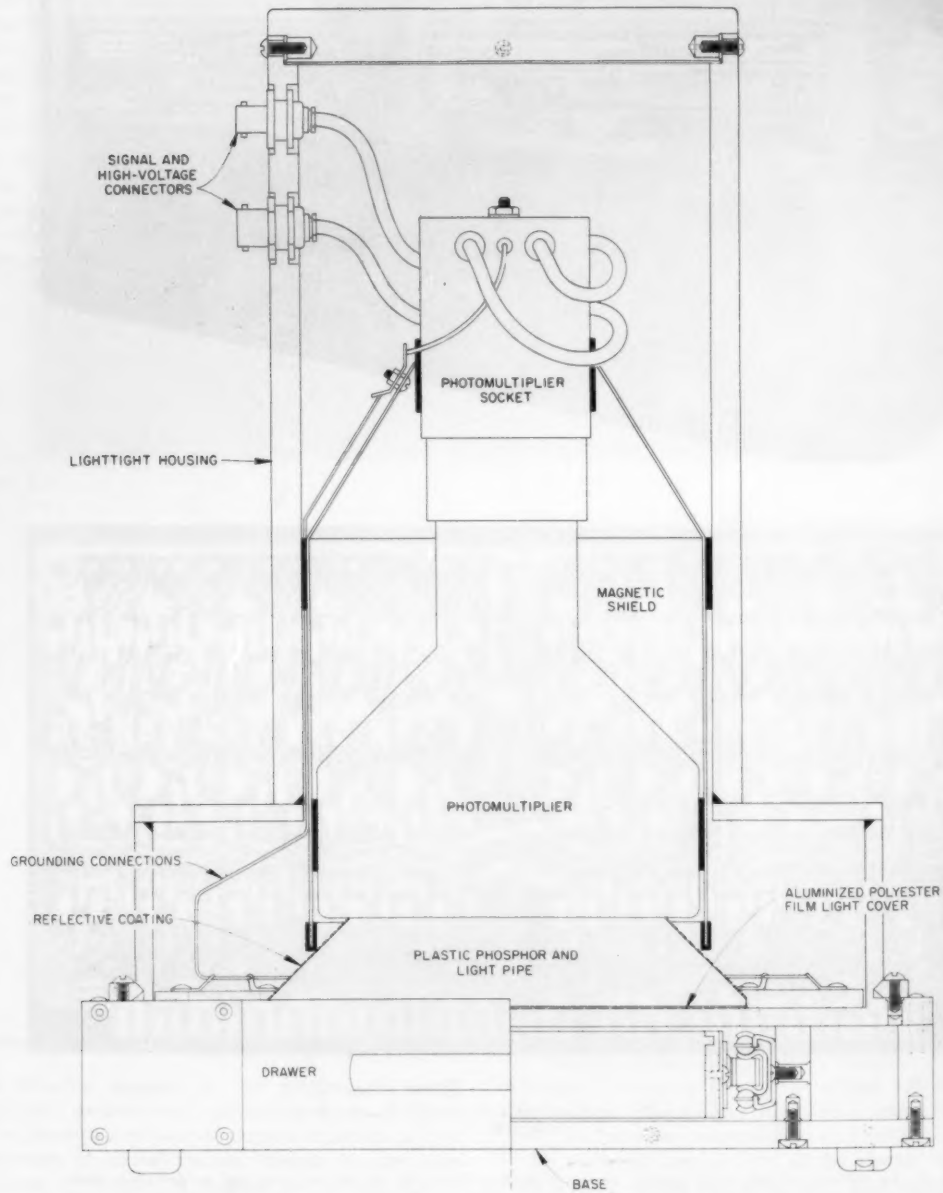

Fig. 4 Cross section of large-area scintillation beta detector.

Previous Tracerlab studies showed the importance of choosing materials of construction with low amounts of natural radioactivity.[5] A new material, polyvinyl chloride, came to our attention; several samples of it were counted, and they showed lower natural background than any conventional materials. Using this material with a very-low-background radioactivity obviated the need for a lead shield. Thus the system was smaller, lighter, and easier to operate.

The circuit of the detector assembly included a standard Dumont type 6364 photomultiplier and a conventional dynode voltage divider. Care was taken to provide electrostatic shielding (a cap on the socket assembly) because the polyvinyl chloride is an excellent insulator. The usual magnetic shield was provided for the photomultiplier, and, because it was conductive, it was also used as part of the electrostatic shield. The electrostatic shielding was completed by connecting the conductive side of the aluminized polyester-film light cover, which protects the phosphor, to ground. The high-voltage and signal leads were brought out through conventional connectors.

In this study the circuit boards were put into a sample receiving drawer in the bottom of the detector (Fig. 5), but a continuous track could have been used on which the boards would pass underneath the detector. If desired, the rate meter can be connected to a recorder; then the serial number of the boards can be written on the chart and a permanent record obtained. This was the technique used during this test. A more sophisticated system could have a digital readout so that the board number and the residual flux count could be printed on a tape.

PROCEDURE

A block diagram of the entire residue-measurement system is shown in Fig. 6. The first step in operating this system was to obtain ^{90}Y tracer from the Brookhaven National Laboratory yttrium generator by elution with 100 ml of 0.5% citric acid which will produce about 80 mc of ^{90}Y with a 1-week interval between milkings. The concentration of ^{90}Y used in this study, 20 mc for 18 gal of flux, is about the minimum that is acceptable for use with the detector signal processed by a rate meter and with the circuit boards of the area used. The tracer was added to the flux, the pumping system was turned on, and the flux was circulated for approximately 1 hr to thoroughly mix the tracer with it. The flux was then sampled with a precision pipet, 0.1 ml being used normally, and stippled on a test board similar to the ones being run. The test board was then dried under a heat lamp and placed in the large-area beta detector and counted. The expected count rate after the wash and rinse cycle of the process was calculated based on the experimentally determined fact that the washing cycle lowers the counting rate by a factor of at least 100. If sufficient activity to ensure a statistically

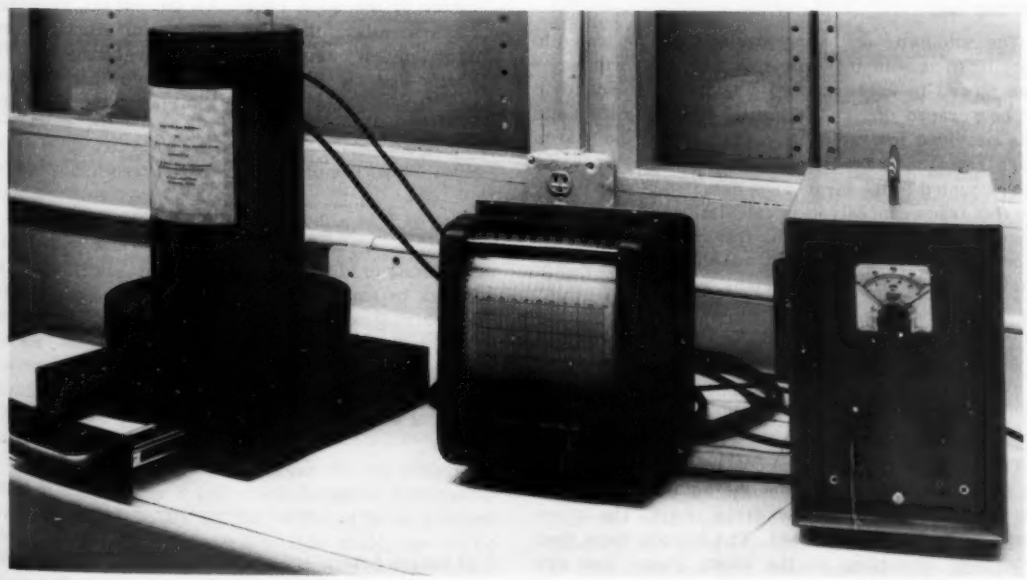

Fig. 5 Large-area beta detector, recorder, and rate meter; drawer of detector open for insertion of circuit board.

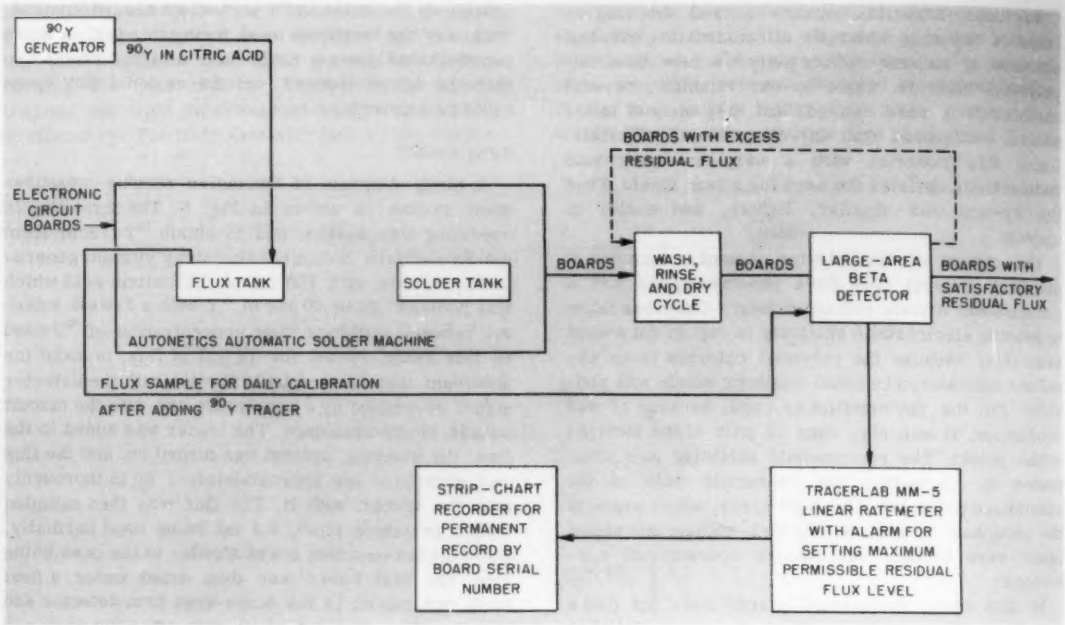
^{90}Y GENERATOR
^{90}Y IN CITRIC ACID
BOARDS WITH EXCESS RESIDUAL FLUX
ELECTRONIC CIRCUIT BOARDS
FLUX TANK
SOLDER TANK
BOARDS
WASH, RINSE, AND DRY CYCLE
BOARDS
LARGE-AREA BETA DETECTOR
BOARDS WITH SATISFACTORY RESIDUAL FLUX
AUTONETICS AUTOMATIC SOLDER MACHINE
FLUX SAMPLE FOR DAILY CALIBRATION AFTER ADDING ^{90}Y TRACER
STRIP-CHART RECORDER FOR PERMANENT RECORD BY BOARD SERIAL NUMBER
TRACERLAB MM-5 LINEAR RATEMETER WITH ALARM FOR SETTING MAXIMUM PERMISSIBLE RESIDUAL FLUX LEVEL

Fig. 6 Block diagram of prototype radioisotope inspection system to measure residual flux on automatically processed circuit boards.

valid count within a reasonable time did not remain after the wash cycle, additional tracer was added to the flux tank.

The automatic soldering machine was operated to specification with the exception that absorbent paper was placed in areas where the tagged flux could drip or be scraped off, and a special disposal receptacle was provided for radioactive waste. After the boards were removed from the soldering machine, several were counted in the large-area beta detector to verify the flux trace concentration calculations. Because the count rate was fairly high, this did not delay the process unduly and allowed washing within 15 min after removal from the machine as specified. Thin plastic place mats were put in the detector drawer before counting any of the soldered boards to prevent accumulation of activity in the detector and thus to prevent a background increase which would reduce counter sensitivity. A background count was made regularly.

The counting rates of the boards were determined as they were removed from the soldering machine and were recorded on the strip chart. The board serial numbers were noted. The boards were then processed according to the wash, rinse, and dry cycle of the specification and again counted. At this time the alarm of the rate meter was set to be actuated if the boards exceeded a precalculated limit based on the calibration of the tracer concentration in the flux and the expected residual flux-removal factor of the wash cycle. As the boards were being counted after cleaning, the serial numbers were noted on the strip chart for a permanent record to verify that the flux residue was below the specification limit. Table 1 shows the counts per minute for

Table 1 RESIDUAL FLUX ON CIRCUIT BOARDS BEFORE AND AFTER WASHING

Net activity, counts/min	
Before washing	After 3-min wash and 3-min rinse
115,000	700
123,000	800
130,000	600
133,000	900
135,000	800
137,000	800
132,000	800
143,000	1100
130,000	600
128,000	400
128,000	600
142,000	600
Av. 131,000	730

10 boards before and after washing. In this particular test only about 0.6% of the flux remained after washing, and the average residue in all the tests was just under 1%.

An earlier investigator[6] of flux residue used ^{36}Cl as a tracer and reported a substantially higher residue (20%) after ultrasonically agitated detergent cleaning. This result does not agree with the residue percentage measured with ^{90}Y tracer. A possible explanation is that chlorine is more chemically active than yttrium and may have exchanged with chlorine or other elements present in the board resin or formed compounds with other elements on the boards.

The validity of ^{90}Y as a tracer for the flux had been established in earlier work by Wright, Fishman, and Smedley[2] who simultaneously compared ^{14}C-tagged glutamic acid with ^{90}Y ion in citric acid as tracers for residual flux. Furthermore, the correlation between the ^{90}Y count rate and the corrosion on boards that had been subjected to the humidity test (Fig. 2) further substantiates the validity of the ^{90}Y tracer.

Personnel Protection and Training

This study was probably the first use of a radioisotope in solution as an industrial production-control tool with production personnel. The health and safety considerations were therefore a necessary part of the system, and careful planning of safeguards in the form of instructions and monitoring instruments was required. The analysis of the safety of the process is first outlined, and this is followed by the training procedures used prior to the start of the test.

SAFETY ANALYSIS

Prevention of overexposure of operating personnel was not difficult because of the low amounts of activity involved.

Previous studies with 80 mc of ^{90}Y in a similar flux tank without a protective enclosure had shown a 2 mr/hr radiation level above the tank; the present study, with 20 mc and an enclosure, showed no radiation increase over background.

The ^{90}Y on individual boards averaged 0.14 μc immediately after soldering, and, since they were handled briefly, no exposure was likely to show on the film badges. A previous study[2] estimated an 8-hr exposure to the operator to be less than 0.5 mr. (Some watches cause more exposure.) The above estimates were verified by the film badges, which indicated no exposure. Disposable gloves were provided for all personnel who were required to handle the boards after soldering.

The amount of residual tracer on the standard-size boards (4 by 8 in.) after complete processing averages about 0.001 μc, as indicated by an average 1050 counts/min per board in this study. As can be seen from the data in Table 1, this amount is not easily removable and therefore should not constitute any hazard. This condition should allow the boards to be handled in the normal manner and not impede the manufacturing process, especially since in 30 days the activity will virtually have disappeared completely.

The air exhaust from the soldering machine was monitored, and, even under the largest rates of circuit-board area possible to process, quantities of ^{90}Y exceeding legal limits do not appear to be exhausted to the atmosphere.

The concentrations of ^{90}Y in the wash and rinse water were very low, and therefore no precaution was taken for its disposition. It was merely allowed to enter the sanitary sewer system of the plant. The required quantity of tracer in the flux was so small (because of the inherently high sensitivity of the detector in the system) that maximum permissible concentration (MPC) levels were easily met by the dilution of the effluent, and the flux was released to the sewer system as it normally would be. The daily average effluent from this plant is 10^6 gal, and the plant operates on a 168-hr week. Because of the low flux concentration and its excellent solubility in water, together with the short half-life of the ^{90}Y tracer, all regulatory personnel involved with the project agreed that the release of this amount of activity would not constitute a hazard. Had there been concern, storage for 30 days would have allowed decay to less than MPC.

Portable survey monitors with thin end-window detectors were used to check the equipment and the machine. A monitor was available at the room exit to check hands, shoes, and clothing of persons leaving. Disposable towels were available in quantity to help keep the work areas clean.

The three classical methods of shielding personnel from exposure—mass, distance, and time—were used: (1) Lucite blocks were placed between the ^{90}Y generator and operators, and the soldering machine was completely enclosed by metal or by glass of approximately equivalent mass. (2) When the ^{90}Y was moved from the radioisotope laboratory to the production area, it was placed in the trunk of the vehicle, so that personnel were kept at a distance, and the soldering-machine enclosure kept personnel away from the tagged flux. (3) All operations were planned and rehearsed, and, although there was no hurry, all critical movements were performed by well-coordinated personnel who were trained to move smoothly and rapidly.

TRAINING

Training of Autonetics personnel started with discussions with their radiation safety personnel. Fortunately they were experienced in the use and handling of radioisotopes and thus needed only specific instructions for the components that were to be used with this study. First, the purpose of this study was explained, and then the individual components of the

study were discussed. The ^{90}Y generator operating instructions state that it should be operated several times after shipping to clear it before actually using any of its product and at least every 3 weeks thereafter to keep the ^{90}Sr contamination low. These preliminary operations of the generator were used as training sessions for Autonetics personnel. Before the operation was attempted, several rehearsals were held using the instruction sheet that was included with the generator when it came from BNL. At the training sessions and at each subsequent operation of the generator, at least two persons were present, one to perform the work and the other to act as monitor and read the step-by-step instructions.

In addition to the radiological health officer, several of the manufacturing-methods personnel, who acted as liaison between this project and the manufacturing department, were trained in problems and functions associated with the project. After the actual production-line use of the radioisotope inspection system was scheduled, the Autonetics radiological safety and production liaison personnel and Tracerlab personnel were introduced to the Autonetics production supervisors, foreman, leadmen, and the operator of the soldering machine. Preliminary discussions outlined the program to them, emphasis being placed on the fact that this was a research study and that their cooperation and contribution would substantially help. They were also told that this study was to be performed so that it would not interfere with their scheduled work.

The final step in the instruction of Autonetics personnel was to hold a briefing session for the operator of the automatic soldering machine, the leadman who was responsible for the work, and the foreman and supervisor of the group. The nature of radioactivity was explained, and the amount of activity that was to be used in the study was related to everyday medical uses of radiation such as dental and chest X rays which have an obviously beneficial aspect. Natural radioactivity occurring as background from both terrestrial and extraterrestrial (cosmic) sources was explained, and examples were given of the change in background radiation from sea level to altitudes comparable to those of Reno, Las Vegas, and Denver. It was pointed out that, because of less atmosphere to provide shielding, the background cosmic radiation in these cities was several times that at sea level.

Since the radioactivity was to be placed in a solution, comparisons were made with naturally occurring radioactivity in solution, the most striking examples of which are certain water wells in Southern Illinois[7] which, while perfectly safe to drink, contain considerably more ^{226}Ra than does the water of other areas. The permissible occupational exposures were also explained, and the total radioactivity to which these personnel would be exposed was shown to be inconsequential. The three basic types of radiation — alpha, beta, and gamma — were also described, and it was explained that the radiation from ^{90}Y was of the beta type. The concentration was described as very low, so that most of the radiation would be self-absorbed in the flux solution. Because the group was small, questions were answered as we went along.

The question of just how much radioactivity would be used per board was answered by showing the radioactivity from a wristwatch with a radium luminous dial with a survey meter. This was compared to the most active of a set of calibration sources with the statement that the activity of each board would be of about that magnitude. The statement was made that, if carrying a watch continuously which contained as much as or more activity than would be on each circuit board was safe and legal, then handling a carrier with two boards on it for a few minutes and also loading the washer to remove the flux were certainly safe. It was also pointed out that the activity on the wristwatch was partly penetrating gamma activity but that on the boards would be relatively nonpenetrating beta activity. The explanations seemed to satisfy the need of the production-line personnel to know that this was indeed a safe project. To record the radiation exposure to personnel, both body and wrist badges were issued to those handling the boards. Personnel required to enter the room but not handle boards were issued body badges, as were the visitors. As expected, the exposure was so small that none was recorded on the badges.

A definite asset to the project was the excellent rapport that the health and safety personnel at Autonetics had established within the entire plant, which ensured that our explanations to the production personnel would carry the weight of the Autonetics management and also allowed us rapid access to services of the plant.

One point that was stressed was that, although the amount of activity that was to be used in this study was relatively small and the hazard from exposure was relatively low, care had to be taken not to ingest any of the activity either through the respiratory system or orally. The explanation was that the glutamic acid hydrochloride in the flux was toxic enough in itself and that, although the amount of radioactivity added to it was definitely a trace quantity, the tracer was an element that tended to lodge and concentrate in bone. And, while the assumption was made that no one would be tempted to drink the flux or any of the water from washing and rinsing the boards, the possibility existed that some slight amount might be carried into a person's system if gloves were not worn and normal sanitary habits of washing before eating were not followed. The operator of the machine was issued heavy rubberized fabric gloves, and other personnel associated with the project were requested to use the disposable gloves.

All gloves used on the project were disposed of as radioactive waste although the room monitor showed a barely detectable residue.

A survey was taken of all personnel normally having access to areas where the tests were performed, and all interested parties were informed so that curiosities were satisfied without disruption of normal work.

Conclusions and Recommendations

This study clearly showed that radioisotopes can be used to quantitatively determine process residues and thus can be used for process control in industry. With proper selection of isotopes, preparation of the facility, slight modification of the operational machinery, and relatively informal training of the production-line workers, radioisotopes can be used in production functions with only small increases in manufacturing costs. In this study the investment in training and room preparation was less than 1 man-week, and the system instrument and ^{90}Y generator costs were substantially less than facility investments of mass-production circuit-board manufacturers. Only a few minutes' additional handling of a circuit board was required to measure the residual flux. Since high standards of cleanliness are already maintained in circuit-board manufacturing, the additional cleanup costs would result only from monitoring and waste disposal.

Many industrial processes require the use of highly corrosive and in many cases toxic or flammable materials, usually fluids. The excellent records of industry in providing for the use of these dangerous materials show that adequate training and supervision already exist. Therefore the addition of radioisotopes to already hazardous materials would result in only incremental costs. The benefits to be derived from the use of radioisotopes as a control tool are improved control of product contamination, resulting in higher reliability; less reworking; and more accurate control. (RHL)

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Measurement of Flow of Liquids and Gases Using Radioactive Isotopes*

By C. G. Clayton†

Abstract: This paper reviews methods of measuring liquid and gas flow in turbulent motion using radioactive isotopes and discusses the principles, limitations, and performance of techniques based on both a constant rate and a sudden injection of radiotracer. Attention is drawn to differences in application to flow measurement in closed conduits and in open channels and streams. Radioisotopes in the form of sealed sources can be used to produce ionization in a gas, which can also be used to measure gas flow. The continuous-ionization method, which provides a means of measuring mass flow, is compared with the mean-velocity method, each of which is used as the basis of instruments for continuous gas-flow measurement.

The measurement of liquid and gas flow using radioactive isotopes is becoming increasingly important as advantages of these methods are more widely appreciated and limitations in the operating characteristics of alternative systems become more apparent as a result of the increasing demand for higher accuracy and reliability over an ever-widening range of flow rates.

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Under controlled conditions, high accuracy and reliability — consistently within ±1% — can be achieved for liquid-flow measurement in closed conduits. The accuracy of measurement of river flow is believed to approach this figure. Errors in volumetric measurement of gas flow based on the velocity method are thought to be much less. Errors in methods based on measurement of sample concentration are probably larger, owing to the difficulty of handling gases; however, further improvement in the accuracy of these techniques is possible.

Radiotracer methods require injection of only small volumes and can be operated with virtually no loss in pressure. Injection and sampling probes can easily be installed in existing or new conduits at very small cost. Measurements can then be made at any time without interrupting the flow. In general, flow-rate measurements that vary over a range of 10^9 can be made using virtually the same equipment. The principal disadvantages of these methods — their discontinuous nature and the relatively short half-life of the tracer that may have to be used — prohibit casual measurements unless a store of activity is available. Therefore radiotracer methods are recommended mainly for flow measurements when highly accurate periodic measurements are requ[illegible] and for calibrating conventional types of continuously operating flowmeters.

All radiotracer methods of measuring the flow of fluids in turbulent motion depend on measuring the velocity or concentration of the tracer at a point sufficiently far downstream from the region of injection for adequate mixing to have been established. The distance required for this is known as the "mixing distance." It is an important factor and may seriously limit the application and ultimate accuracy of measurement.

Methods based on induced ionization are also being developed for gas-flow measurement. These methods operate continuously without impeding the flow and can be used to measure mass flow.

Several systems of flow measurement and control based on the mechanical movement or rotation of a vane have been improved by using a radioiostope as the indicating element.

Principles of Radiotracer Methods of Measuring Turbulent Flow

The use of radioactive isotopes to measure the flow velocity of liquids in pipes and open channels was first mentioned by Joly[1] in 1922. In general, the methods are very similar to the chemical tracer techniques, but the use of radioactive isotopes offers certain advantages (mainly on account of the sensitivity of radiation detection) and results in differences in operating procedure, which reflect on the accuracy and convenience of the methods.

CONSTANT-RATE-INJECTION METHOD

Flow measurement by the constant-rate-injection method is based on a comparison between the concentration, C_1, of a tracer that is continuously introduced at a known rate q with the concentration, C_2, of samples removed at some position beyond the mixing distance. The rate at which activity is injected is equal to the rate at which it passes the sampling point:

$$qC_1 + QC_0 = (Q + q)C_2 \qquad (1)$$

where C_0 is the initial concentration in the stream, which is flowing at the rate Q. Hence

$$Q = q(C_1 - C_2)/(C_2 - C_0) \qquad (2)$$

Generally, $C_1 \gg C_2$ and $C_2 \gg C_0$, and Eq. 2 reduces to

$$Q = q(C_1 / C_2) \qquad (3)$$

Thus the flow rate can be determined absolutely by comparing the concentration of the injected solution with that of samples removed from the flow. Provided the flow is turbulent, the method is completely independent of fluid velocity, conduit dimensions, or any variations in these dimensions. Since only the concentration of the tracer is required at the sampling point, the samples can be taken from a branch or standpipe that leaves the main pipe after lateral mixing has occurred if the main pipe itself is inaccessible. The flow velocity in the branch pipe is immaterial and may be laminar. This important result has many applications in flow measurement in industrial plants.

Since for most applications (including flow rates up to 200 cu ft/sec for liquids) an injection time of less than 30 min is usually sufficient to establish uniform concentration at the sampling point, a unidirectional pump in the form of a piston moving in an accurately machined cylinder can be used. A well-proved injector for liquid-flow measurement, shown in Fig. 1, has a measured error in the injection rate of less than ±0.05%.

A small volume of the injected solution is obtained before and after injection and later used to determine C_1.

The samples from the pipe can be collected in any suitable container, and the concentration C_2 is then compared with that of a known dilution of specimens of the injected solution (C_1) in the same high-grade counting system. The concentration of the aliquot used to obtain C_1 is best made about equal to C_2 (for approximate equating of counting rates); hence very high dilution factors, e.g., up to 10^7, may be necessary. Since the accuracy of flow measurement is directly proportional to that of measuring C_1, great care must be exercised in making the dilution.

The choice of detector depends on the preferred isotope. When ^{24}Na is used for water-flow measurement, a reentrant measuring container with a volume of about 2.5 liters has been found convenient to handle and gives acceptable counting efficiency.

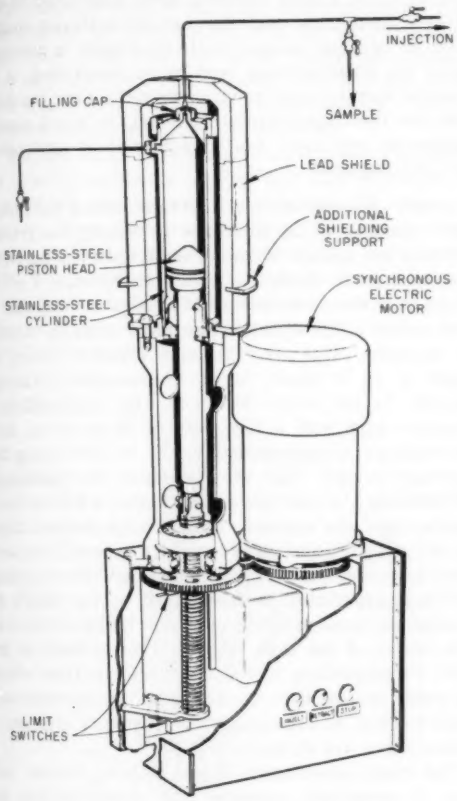

Fig. 1 Constant-rate injector.

SUDDEN-INJECTION METHODS

In the sudden-injection method a volume, V_1, of solution with concentration C_1 is rapidly injected into the stream, and the concentration is examined at a point beyond the mixing distance. Equating the activity injected with the total in the fluid gives

$$(C_1 - C_0)\ V_1 = \int_B \int_L (C - C_0)\ dL\ dB \qquad (4)$$

where C_0 = initial concentration in the stream (assumed to be constant)
B = lateral coordinate
L = longitudinal coordinate
C = concentration in the elemental volume $dL\ dB$ at any instant of time t

The longitudinal flow velocity through this element is $v = dL/dt$, and Eq. 4 can be written

$$(C_1 - C_0)\ V_1 = \int_B \int_t (C - C_0)\ v\ dt\ dB \qquad (5)$$

Now, if the concentration $(C - C_0)$ is evenly distributed over an entire cross section (i.e., uniformly independent of B) and if the velocity, v, over that cross section is unchanging (i.e., independent of t), Eq. 5 can be integrated separately with respect to B to give

$$(C_1 - C_0)V_1 = \int_B v\ dB \int_t (C - C_0)\ dt = Q \int_t (C - C_0)\ dt \qquad (6)$$

where Q is the total flow rate. If $C_1 \gg C_0$, as is usually the case, and since $V_1C_1 = A$, the total activity of the radioisotope introduced, the flow rate is given by

$$Q = A / \int_t C\ dt \qquad (7)$$

Thus, to determine the flow rate, it is simply necessary to know the total injected activity of the isotope and the value of the integral.

Continuous-Sample Method. The integral may be evaluated in two ways, either of which is the basis of the continuous-sample method. In one way the concentration at the sampling point is measured continuously, a concentration–time curve is constructed, and the integral is evaluated numerically. However, since the concentration is changing smoothly, a strictly continuous measurement is not necessary, and the curve is derived by measuring the concentration of discrete samples removed periodically. Alternatively, the integral is evaluated by using the relation $\int_t C\ dt = \overline{C}(t_2 - t_1)$, where $\overline{C}$ is the mean concentration of samples removed at equal time intervals. In practice, $\overline{C}$ may be determined from a single measurement of the concentration of samples mixed in a suitable container.

The principal advantages of the continuous-sample method over the constant-rate-injection method are:

1. It is economical in equipment since apparatus for a continuous, sustained injection is not required.

2. It is less sensitive to the position of the sampling point. In the constant-rate-injection method, the sampling point is restricted to a region between the mixing distance and a distance at which the longitudinal dispersion begins to limit the time during which the concentration remains constant. In the continuous-sample method, this latter requirement does not apply, and the only consideration is that the concentration should be measurable. This difference becomes important in pipelines when the measuring section includes valves, bends, and other devices that increase dispersion and tend to destroy the region of uniform concentration.

3. If samples removed during the transit period of the tracer are mixed, a single concentration measurement is sufficient to determine the flow rate. Samples are taken throughout the entire time that the tracer is passing the sampling point, and there is no delay for the concentration to rise to a steady value. Hence, for similar measured concentrations, less total activity has to be injected.

4. Only an approximate knowledge of the tracer distribution is required. Samples must be removed before the tracer arrives, but sampling may be continued until well after the estimated transit time although too many of these samples will reduce the concentration in the mixture to be measured.

Although neither the constant-rate-injection nor the continuous-sample method is a direct recording system since samples have to be removed for assay, a rapid measurement and result are possible in many applications.

Total-Count Method. If a counter is considered to be in position in the flow at the sampling point in the continuous-sample method, the number of counts, N, recorded during the passage of the tracer is given by

$$N = F \int_t C\,dt \quad \text{or} \quad \int_t C\,dt = N/F \tag{8}$$

where F is a constant that depends on the detector efficiency. Inserting this in Eq. 7, we have

$$Q = AF/N \tag{9}$$

which is Hull's equation[2] for the total-count method.

In practice, F is obtained by calibrating the detector, and, since this must be done under the same geometry conditions as the flow measurement, it is more convenient not to immerse the detector in the flow but to define a reproducible geometry by using a reentrant chamber through which the fluid from the main pipe passes at a constant rate. This chamber can be calibrated with an aliquot of the injected solution.

It is worth noting that the specific activity of the injected solution does not have to be known. If an aliquot of the injected solution (concentration C_1) is used to calibrate the detector, the aliquot concentration is C_1/f where f is the dilution factor. If this sample is counted for time T, to give a total count n, then $n = F(C_1/f)T$. And, since $A = V_1C$, where V_1 is the volume of injected solution, $AF = V_1C_1F = V_1(nf/\mathrm{T})$. Then from Eq. 9

$$Q = (V_1 f/T)(n/N)$$

so that the determination of Q depends only on the ratio of two counts, the counting time of one of them, a dilution factor, and the volume of injected solution.

The principal advantage of the total-count method is that an instantaneous reading is obtained, and, if the calibration factor, F, has been derived previously, the system can be arranged to measure flow rate directly. The principal disadvantages are the low statistical accuracy that may be present in N, which results from a total counting time restricted to the pulse transit time, and the need for detecting equipment to operate reliably while the tracer is passing. Since the disadvantages tend to be overriding, it is doubtful that the total-count method can ever compete with the continuous-sample method, in which higher statistical accuracy can be achieved by prolonging the counting time.

Isotope-Velocity Method. In this method the linear flow velocity, v, is measured by timing the tracer between two points of known separation and deriving the volumetric flow, V, from the relation $V = vA$, where A is the cross-sectional area of the channel.

In practice, the injection should be made as rapidly as possible. This may be done within 10 msec, in pipes of up to about 30 in. in diameter containing liquids, by the device shown in Fig. 2. In a 20-in.-diameter pipe with a flow rate of 20 cu ft/sec, this corresponds to approximately 1 in. of flow during the injection period. The two detectors are positioned downstream, beyond the mixing distance for the particular injection system. The distance between them is not critical, although accuracy is generally highest when the detectors are well separated. The smallest practical separation is determined by the ability to distinguish between the traces from the two detectors. The center of the area of each trace is taken as the point corresponding to the mean velocity from which the pulse transit time is calculated. An approximate, rapid method of calculating the center of area and a typical trace are shown in Fig. 3.

The chief advantages of the velocity method are that it conserves activity, the detectors can be mounted outside the pipe, and the monitoring system can be arranged to read flow rate directly. The velocity method is very convenient to operate, and repetitive measurements can be made readily. The chief disadvantages for volumetric flow measurement are the dependence on an accurate knowledge of the cross-sectional area and the sensitivity to structures such as valves and orifice plates, which increase the longitudinal dispersion of the tracer to an extent that it is no longer measurable.

Liquid-Flow Measurement

CLOSED CONDUITS

Choice of Radioisotope. The choice of a radioisotope for flow measurement depends mainly on the solubility and stability of the tracer in the flowing fluid, radioactive half-life, type and energy of radia-

tions emitted, maximum permissible level in solution in case of ingestion, and maximum specific activity available.

The first requirement is common to all tracer methods—not just to one using radioactivity. However, the problem of stability is often more stringent with radioisotopes owing to the high sensitivity of the radiotracer methods in terms of the weight of material used. Care must be taken to avoid precipitation and wall adsorption; additional nonradioactive material may have to be injected simultaneously. The total injected weight is still small, however.

For the other requirements a compromised choice is usually necessary. A long half-life is more convenient for storage and measurement but should be avoided from the viewpoint of health hazard, especially if there is possibility of drinking-water pollution. For small-scale investigations it may be possible to use a short-lived daughter isotope, e.g., ^{132}I ($T_{\frac{1}{2}}$ = 2.26 hr), which can be prepared on site from the 77-hr parent ^{132}Te.

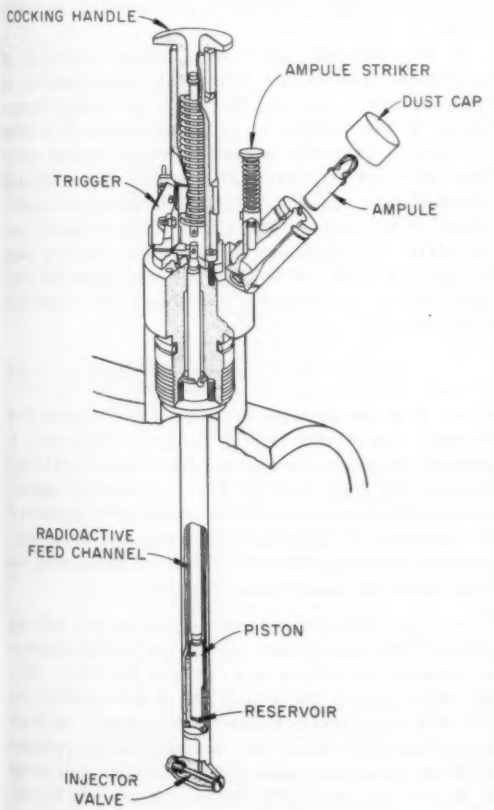

Fig. 2 High-speed injector for measuring liquid flow by the isotope-velocity method.

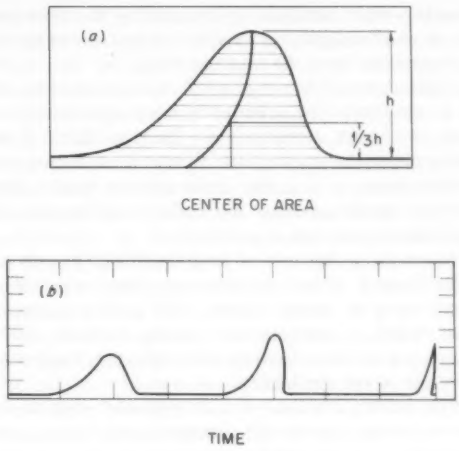

Fig. 3 (a) Method of estimating center of area of pulse, and (b) typical trace in the isotope-velocity method of measuring flow.

Sensitivity is highest when large volumes of liquid are measured and the tracer is a high-energy gamma emitter, but such nuclides are difficult to transport in high activity. A pure beta emitter is more easily handled, but the limited counting volume results in a low measurement sensitivity. When sampling techniques are used, the sensitivity can be increased by using quantitative chemical extraction. With the velocity method high-energy gamma radiation is essential for operating detectors outside the pipe.

Estimation of Mixing Distance and Dispersion. At present there is no complete, experimentally verified theory by which the mixing length in a given system can be predicted. The effects of changing the Reynolds number and the decrease in mixing length resulting from the use of multiple-injection systems, for instance, can be established only qualitatively. Some experimental results are available, however. Bonnin, Dumas, and Lievre[3] studied the effect of injecting salt solution on the pipe axis and at various other positions over the cross section of a pipe containing fluid with a Reynolds number between 10^4 and 10^5. However, they used solutions with very high salt concentrations, so that density effects were appreciable, and therefore the results are to be accepted with some caution.

The curve labeled "center injection" in Fig. 4 indicates[4] how a radioisotope injected from a single orifice on the axis of a pipe of 10.32 cm diameter approaches uniform concentration for a flow corresponding to a Reynolds number (Re) = 7.7×10^4. The curve labeled "multiple-edge injection" shows the advantage to be gained from using four injection orifices equally spread around the pipe wall. These

results were obtained by comparing the concentrations of 10 samples at each of several cross sections downstream from the injector plane.

The approach to a regime of uniform concentration is demonstrated by calculating the progressive variation of χ^2 with distance down the pipe. Each χ^2 value determines the probability, p, that a repetition of the observations at a given cross section would show a greater deviation from the mean concentration than that observed in the experiment.

Apart from the use of long lengths of pipe to promote mixing solely by turbulent dispersion, structures such as bends, valves, and orifice plates are also useful in reducing the mixing distance, but reliable quantitative information indicating their effectiveness is not available.

The mixing distance can be reduced considerably by including circulating pumps in the test section. Unfortunately the mixing characteristics of pumps are not at present generally available, and further information is required to establish the performance in terms of an equivalent length of straight pipe. Table 1 shows the variation in concentration measured over a cross section in the output of a mixed-flow pump after a single wall injection[5] and indicates the benefit to be gained from using a pump to decrease the mixing distance. Turbines, condensers, and reaction vessels may also be included in the test section, but, if the longitudinal dispersion is large, the activity required must be increased correspondingly.

Table 1 VARIATION IN RADIOISOTOPE CONCENTRATION OVER A 20-IN.-DIAMETER CROSS SECTION IN A MIXED-FLOW PUMP OUTLET AFTER A SINGLE WALL INJECTION IN THE INLET

Sampling-point distance from pipe center, in.	Total recorded count	Percentage of mean concentration
	Set 1*	
7.56	37,360	102.2
5.56	36,590	100.1
3.56	36,406	99.6
0.56	36,582	100.1
2.47	35,876	98.2
4.47	36,596	100.1
6.47	36,482	99.7
	Set 2*	
7.53	32,136	100.3
5.53	32,570	101.7
3.53	31,928	99.7
0.53	31,330	97.9
2.44	31,754	99.2
4.44	32,148	100.4
6.07	32,288	100.8

*The two sets correspond to two diameters at right angles.

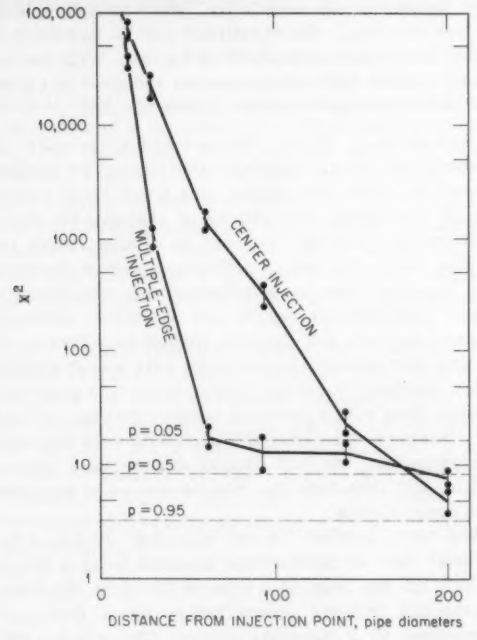

Fig. 4 The χ^2 values for sets of 10 samples taken across vertical and horizontal diameters. Tracer is considered mixed when p is between 0.05 and 0.95.

In the methods based on a sudden injection of tracer, a time during which the concentration is constant never occurs. However, the same dependence of the position of sampling points on mixing distance is generally accepted. With a sudden injection the total dispersion also affects the maximum concentration and the extent of longitudinal distribution of the tracer. The effect of the hydraulic parameters on dispersion in a straight circular pipe has been worked out by Taylor,[6] who finds the concentration at a distance x and time t from injection to be given by

$$C(x,t) = At^{-\frac{1}{2}} \exp\left[-(x - ut)^2/4kt\right] \qquad (10)$$

where A is the injected activity, u is the mean flow velocity, and k is a virtual diffusion coefficient. In general, Taylor's theory has been reasonably well verified for long straight pipes containing incompressible fluids, although data collected by Sjenitzer[7] for commercial overland pipes indicate some discrepancy between observed values of dispersion and those predicted from Taylor's theory.

Practice. The constant-rate-injection and velocity methods have been widely used in the United Kingdom to measure flow rates up to 260 cfs ($Re = 2.5 \times 10^6$), and the accuracy and reliability of the methods are now well established. Earlier publications[8,9] indicate the accuracy to which both systems can be operated in 20-in.-diameter pipes with controlled flow rates up to 20 cfs ($Re = 10^6$). More recent results, referred to a weighbridge measurement (accuracy ±0.4%) and obtained in an experimental 4-in.-diameter pipe for $Re = 7.7 \times 10^4$, are given in Tables

2–4. The data in Table 2 for the velocity method indicate that the reproducibility of this measurement is as good as that of the weighbridge. The results in Tables 3 and 4 were obtained from two cross sections at 150 and 200 diameters distance from the injection point and demonstrate the small variations in concentration that can be expected in a series of samples used to measure flow by the constant-rate-injection method. The low values of standard error are remarkable when compared with results quoted for other methods of flow measurement and are accountable almost solely in terms of counting statistics.

When the two methods are used to measure the flow of condenser cooling water in power stations, the consistency of results shown in Tables 2–4 is rarely obtained, mainly because of variations in head across the pump (when using tidal water, for example). When transient variations are present in the stream, the two techniques respond differently. A measurement by the velocity method is an average over the transit time between detectors. Apart from an effect due to longitudinal dispersion, if the sample is collected rapidly, the constant-rate-injection method gives an almost instantaneous measurement of flow referred to the injection point. An average value can be obtained by suitably increasing the collection time. Results like those shown in Tables 3 and 4 give confidence to the conclusion that variations in flow obtained in practice are inherent in the system and not in the measurement.

Table 2 FLOW MEASUREMENT BY THE VELOCITY METHOD ON A 4.16-IN.-DIAMETER PIPE AT Re = 7.7×10^4

Test	No. of measurements	Flow rate, lb/sec: Weight method	Flow rate, lb/sec: Velocity method	Error, %	Standard deviation (σ)	Coefficient of variation ($100\ \sigma/\chi$)
1	20	18.12	18.25	+0.71	0.0057	0.3
2	14	18.14	18.26	+0.66	0.012	0.8
3	16	17.55	17.56	+0.06	0.017	0.3
4	13	17.47	17.51	+0.23	0.024	0.5

Table 3 VARIATION IN CONCENTRATION (CORRECTED COUNT) IN THREE GROUPS OF 10 SAMPLES REMOVED AT 150 DIAMETERS DOWNSTREAM FROM THE INJECTION POSITION IN A 4.16-IN.-DIAMETER PIPE CONTAINING WATER WITH Re = 7.7×10^4

Sample	Corrected count	Percentage of mean	Sample	Corrected count	Percentage of mean	Sample	Corrected count	Percentage of mean
1	171,346	99.7	11	171,015	99.5	21	171,285	99.6
2	172,415	100.3	12	171,902	100.0	22	171,419	99.7
3	171,895	100.0	13	172,178	100.1	23	171,017	99.5
4	172,762	100.5	14	172,434	100.3	24	172,362	100.2
5	172,242	100.2	15	172,105	100.1	25	172,950	100.6
6	171,315	99.6	16	172,053	100.0	26	172,478	100.3
7	171,826	99.9	17	171,950	100.0	27	172,099	100.1
8	172,460	100.3	18	172,253	100.2	28	171,930	100.0
9	171,917	100.0	19	171,773	99.9	29	172,220	100.2
10	171,202	99.6	20	171,898	100.0	30	172,120	100.1

	Samples 1–10	Samples 11–20	Samples 21–30
Mean corrected count*	171,938	171,956	171,988
Standard deviation	534	383	592
Relative standard deviation	0.31%	0.22%	0.34%
Standard error of mean	169 (0.098%)	121 (0.070%)	187 (0.109%)

*The different mean values of corrected count compared with Table 4 are due to the use of a higher efficiency detector.

Table 4 VARIATION IN CONCENTRATION (CORRECTED COUNT) IN THREE GROUPS OF 10 SAMPLES REMOVED AT 200 DIAMETERS DOWNSTREAM FROM THE INJECTION POSITION IN A 4.16-IN.-DIAMETER PIPE CONTAINING WATER WITH Re = 7.7×10^4

Sample	Corrected count	Percentage of mean	Sample	Corrected count	Percentage of mean	Sample	Corrected count	Percentage of mean
1	162,611	99.5	11	163,458	100.2	21	163,596	100.3
2	163,486	100.1	12	163,352	100.1	22	162,596	99.6
3	162,928	99.7	13	163,261	100.0	23	162,166	99.4
4	162,898	99.7	14	163,168	100.0	24	163,806	100.4
5	163,671	100.2	15	162,889	99.8	25	163,188	100.0
6	163,570	100.1	16	162,763	99.7	26	163,615	100.3
7	163,196	99.9	17	163,014	99.9	27	162,847	99.8
8	164,111	100.5	18	163,493	100.2	28	163,741	100.3
9	163,350	100.0	19	163,603	100.2	29	163,322	100.1
10	163,862	100.3	20	162,999	99.9	30	162,890	99.8

	Samples 1–10	Samples 11–20	Samples 21–30
Mean corrected count*	163,368	163,200	163,177
Standard deviation	466	278	543
Relative standard deviation	0.29%	0.17%	0.33%
Standard error of mean	147 (0.090%)	88 (0.054%)	172 (0.105%)

*The different mean values of corrected count compared with Table 3 are due to the use of a lower efficiency detector.

If the cross-sectional area of the measuring section is known only approximately, the velocity method cannot be used, and for high accuracy either the constant-rate-injection or continuous-sample system must be used. However, although established as an accurate method, at high flow rates the constant-rate-injection method is often inconvenient because of the amount of activity required and the time needed to prepare the injection solution. There is also a delay before results are finally worked out. Under these circumstances, when a long series of measurements has to be made on one system, it is convenient to operate the constant-rate-injection and velocity methods together over a time when the flow rate is constant and then to continue the series using only the velocity method. This approach amounts to calibrating one method with the other in order to derive a value for the cross-sectional area for use in subsequent measurements by the velocity method. In this combined system of calibration and measurement, the accuracy of quoted results is tied to the accuracy of the derived value for the cross-sectional area, and this may be checked subsequently by direct measurement of the internal dimensions of the conduit.

When this technique was first tried during measurements of the cooling-water flow at Uskmouth "B" power station, the results were quoted on the basis of a derived value for the cross-sectional area of 24.84 ± 0.17 sq ft, which may be compared with a measured value of 25.1 ± 0.1 sq ft.

Comparable figures establishing the accuracy of the continuous-sample and total-count methods have not been published, although Hull[10] considers that 1% is the limit of reliability of the total-count method and a theoretical limit of 0.1% should be attainable with the continuous-sample method.

The application of radioisotope techniques to a wide variety of measurement problems in industrial plants has not yet been fully exploited in the United Kingdom, even though a wide range of investigations has been reported in the literature. A recent investigation[11] in which the velocity method was used to measure the flow in boiler tubes in a power station in the United Kingdom illustrates the flexibility of these techniques. At the input end the tubes were known to contain water at a temperature of 285 °C at a pressure of 1000 psi, while at the output end, owing to an increase in temperature, steam was also present. To measure flow in the tubes, 20 ml of water containing about 2 mc of ^{24}Na was injected into the system, and the transit times were observed by using three water-cooled scintillation detectors mounted on the outer side of each pipe so that the flow in adjacent sections could be measured. By comparing the transit times in adjacent sections in a number of pipes over times of several hours, variations in water flow rate as well as variations in steam formation rates could be demonstrated. A second experiment carried out after the inner walls of the pipes had been cleaned with acid showed a marked decrease in flow variations in each pipe.

OPEN CHANNELS AND RIVERS

Since in most open channels and rivers the cross section is not uniform and the cross-sectional area may vary because of changes in hydraulic gradient, the velocity method is only rarely applicable. The other methods described above are widely used, and the relative merits of each for open-channel measurements have been discussed elsewhere.[12]

Tracer Adsorption. One of the most important differences between flow measurement in pipes and open channels or rivers arises from the increased possibility of tracer adsorption, by both suspended material and material in the bed, and this tends to impose a practical limit on the length of the test section.

The adsorption of radioactive isotopes in very dilute solutions has been studied in connection with the use of tracers to follow underground water movement, and it has been found that, in general, anions are adsorbed less than cations.[13] Suitable tracers include Br^-, I^-, and complex compounds such as chromium EDTA. Cationic tracers can be used if the river already contains cations of the same type so that loss of radioactive tracer is decreased. Since Na^+ normally exists in trace amounts in most rivers, ^{24}Na is a satisfactory tracer.

The problem of adsorption is overcome completely by the use of ^3H, which is unique as a tracer for water-flow measurement. Only an isotope effect could account for its partial removal from water. It is available in high specific activity and is not expensive. In spite of its long radioactive half-life, it is one of the least toxic isotopes, the maximum permissible concentrations in drinking water being 0.03 μc/ml for occupationally exposed workers (168-hr week) and 0.001 μc/ml for the general public. Tritium emits beta particles with 18 kev maximum energy and hence requires specialized laboratory equipment for accurate measurement. It cannot be measured directly in the river, so the total-count method is not applicable, and its presence can be followed directly only by simultaneous use of a gamma emitter or a visual tracer.

Estimation of Mixing Length. Several attempts have been made to calculate the mixing length in open channels.[14–16] Assuming that vertical mixing is much more rapid than transverse, Rimmar[15] considered the problem as a two-dimensional eddy-diffusion problem and found that the mixing length is given (in meters) by

$$L_{mix} = 0.13(B^2/H)(C/g)(0.7C + 6) \quad (11)$$

where H = depth of stream, meters
B = breadth of stream, meters
g = gravitational constant, MKS units
C = Chezy coefficient

Usually C is between 15 and 50, depending on the nature of the bed.

Assuming that transverse and longitudinal mixing coefficients are equal and that the dispersion is a true Gaussian function, Hull[16] found the minimum distance for complete mixing to be given by

$$L_{mix} = 50Q^{1/2} \quad (12)$$

where Q is the total mass flow, in cubic meters per second. In the derivation of Eq. 12, center-point injection was assumed and no account was taken of frictional drag.

Neither of the above equations has yet been shown to agree with experimental observations, probably because of the widely varying cross sections that are found in practice and the difficulty in estimating the Chezy coefficient.

In open streams a rough visual estimate of the mixing length may be made by introducing a concentrated dye solution at the injection point and observing the general spread up to the measuring point. Alternatively, two radiation monitors may be used: at the center of the channel and close to one side. The mixing length is exceeded when the detector readings are equal. This technique is particularly useful when measuring the flow of rivers in which stagnant zones are present in the measuring reach because it gives a useful qualitative estimate of the mixing length.

Once the mixing length has been established, samples may, in principle, be removed from any point further downstream. In practice, however, in a natural stream or artificial open channel, a decrease in flow may occur due to seepage, and an increase in flow may arise from surface or groundwater. Seepage that occurs between the mixing length and the sampling point will not affect the concentration of tracer and will therefore go undetected. Surface water and groundwater, if completely mixed at the sampling point, will be measured directly. However, for accurate measurement, a test section should be chosen in which groundwater accretion and seepage are both negligible.

Gas-Flow Measurement by Radiotracer Method

Although the methods described above for measuring liquid flow rates can, in principle, be used equally well to measure gas flow, there are a number of important differences in the practical application of the methods. Mixing distances in gases have been calculated for single, annular, and moving-point injectors by Jordan,[17] who showed reasonably good agreement between theoretical and experimental results in the case of a radioactive gas continuously injected into the flow. Discrepancies between theoretical and experimental results for the longitudinal dispersion of a tracer rapidly introduced into the flow were observed by Clayton, Webb, and Whittaker[18] and by Hodkinson and Leach.[19]

CHARACTERISTICS OF GAS-FLOW MEASUREMENT

Because of the low density of gases and the fact that gas pipes tend to be of smaller diameter than pipes containing liquids, the linear velocity of gases in turbulent motion is higher than that of liquids. Typical velocities are 0.1 to 10 ft/sec for liquid flow and 10 to 1000 ft/sec for gas. The application of the velocity and total-count methods is particularly affected — increased activities and faster recording systems are required. Further, the greater difficulty of sampling gases as compared to liquids limits the application of the constant-rate-injection and continuous-sample methods.

The compressible nature of gases generally increases the difficulty of designing a suitable injection system. When the constant-rate-injection method is used, great care is required to guarantee an accurate and constant injection rate. In the sudden-injection methods the usual system of injecting the tracer under high pressure through a small orifice is unsuitable for fast injection since compression ratios exceeding about 10/1 may produce sonic velocities and shock waves at release. A design of an injector[20] that overcomes this difficulty is shown in Fig. 5. The main feature of this instrument is a piston that contains two cylindrical gas chambers at right angles to the main axis of the piston. One chamber forms part of the main supply line in which the flow is to be measured, and the second chamber is filled with a radioactive gas from a reservoir. For injection the piston is operated by a compression spring so that the chamber containing the radioactive gas forms part of the main pipe. In this way the gas is injected with little disturbance in about 2 msec. If the diameter of the main pipe exceeds the injector-chamber diameter, it is usually convenient to mount the injector in a short branch through which gas can be passed at a pressure slightly above that of the gas in the pipe and for a time long enough to ensure that the radioactive gas becomes part of the main flow.

Because of compression the linear velocity of a gas flowing in a pipe may not be constant, and a correction may be necessary when the velocity method is applied.

In the sudden-injection methods, the length of pipe occupied by the tracer may be increased by gaseous expansion as well as by turbulent dispersion. The proportion of tracer exposed to a detector at any moment is therefore decreased, and this decrease, which is calculable,[18] must be included in an assessment of the tracer activity needed to carry out the measurement.

The number of radioactive isotopes that can be used as gaseous tracers is limited compared with the number available for liquid-flow measurement. The nuclides ^{222}Rn, ^{133}Xe, and ^{41}Ar have often been used,[21–23] but their short half-lives are invariably a disadvantage. Methyl bromide labeled with ^{82}Br has the advantage of high-energy gamma emission and a half-life of 35.9 hr, but the highest available specific activity (110 mc/liter) limits the range of application. Krypton-85 is the most commonly used radiotracer for gas-flow measurement. Although it is mainly a beta emitter (0.67- to 0.52-Mev gamma rays emitted during 0.4% of the total disintegrations), its long half-life (10.6 years), high available specific activity (80 curies/liter), and low toxicity override any disadvantages. In spite of these difficulties, tracer methods of gas-flow measurement have been used successfully in a large number of applications.

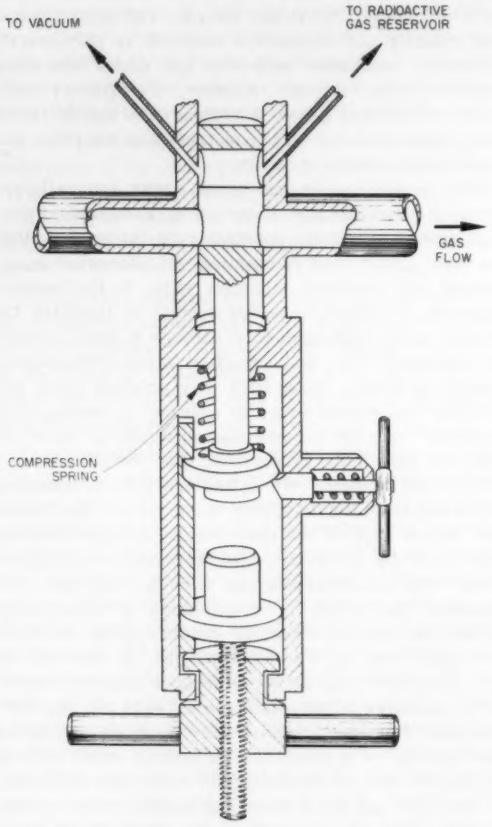

Fig. 5 High-speed injector for radioactive gases. When the trigger is released, the spring drives the chamber, which is filled with a radioactive gas, into the main stream, replacing the section normally present.

USES

The choice of tracer techniques to measure gas velocities is particularly attractive in two-phase systems, such as in the pneumatic transport of dust particles, since independent measurement of gas and particle velocities is difficult, if not impossible, by pressure-difference devices. By using radioisotopes as tracers, the gas and solids fractions can be labeled independently, and relations between driving-pump speed, particle size distribution, particle velocity, gas velocity, gas pressure, and solids density, for example, can be established readily. An experiment of this kind has recently been carried out by Berry and Webb[24] on a closed-cycle graphite-dust suspension loop using ^{133}Xe and ^{56}Mn as the radiotracers. Depending on the dust concentration (0.02 to 0.39 lb/cu ft), it was shown that differences of up to 50% existed between the dust and gas velocities, while a Venturi meter calibrated in terms of average fluid density closely followed the dust velocity.

The use of the total-count method to measure gas flow rates varying from a few to 10^5 scfm has recently been reported by Fries.[25] Krypton-85 has been used in calibrating meters and in measuring flow in unmetered systems and where local pressure changes preclude other methods, as at the air intake of internal combustion engines. Quoted accuracies lie between 1 and 3%. In one application in a synthetic ammonia plant, methane flow was measured by the total-count method to determine whether observed variations in methane yield could be attributed to inaccuracies in the orifice flowmeter. Krypton-85 tracer was injected into the pipe containing methane at ambient temperature and 10 atm pressure over a time of 10 min, and the orifice plate was located midway between injection and sampling points. The total count was measured using a Geiger counter mounted in a reentrant chamber through which a fraction of the main gas stream was passed at a rate of about 1 cfm. It was concluded that the total-count results agreed with the orifice plate within 3% over the required flow range, and the observed difference in yield could therefore not be attributed to inaccuracies in the orifice plate meter.

The use of the continuous-sample method to measure a range of steam flow rates has also been reported by Fries.[26] A few milliliters of tritiated water containing about 5 mc of activity was injected for

each 10,000 lb of steam in the system. The tracer was injected over a period of a few seconds with a pressurized container, and it was assumed that the water vaporized and mixed with the steam rapidly after injection. A sample of the steam was obtained through a sampling line, passed through a small condenser, and collected in a graduated cylinder (Fig. 6). Results for one experiment are given in Table 5.

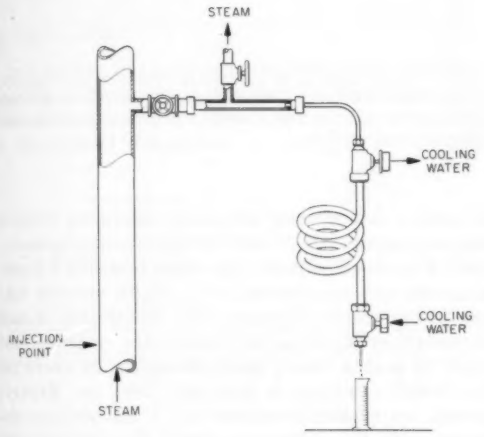

Fig. 6 Steam-flow measurement by the continuous-sample method, showing arrangement for collecting sample.

Table 5 RESULTS OF MEASURING STEAM FLOW BY THE CONTINUOUS-SAMPLE METHOD[26]

Test	Pressure, psig	Temp., °F	Flow rate, lb/hr: Plant flowmeter, corrected for pressure and temp.	Continuous-sample method: At 225 ft	Continuous-sample method: At 1100 ft
1	132	368	11,600	12,200	12,500
2	133	394	11,600	11,700	11,400

Gas-Flow Measurement by Induced Ionization

The measurement of gas flow using ionization produced by radioisotopes dates from 1928 when Blake[27] first suggested the continuous-ionization method. Since then several patents have been granted on more sophisticated systems, and a number of papers have been published dealing with various design features. However, there is very little information on the operational performance of instruments of this type. In view of the demand for greater accuracy of gas-flow measurements, especially at high velocity, and the interest in direct mass-flow measurement without introducing pressure loss, more instruments of this type will probably be used in the future.

CONTINUOUS IONIZATION

In this method ions are produced continuously by either an alpha- or beta-emitting source and are collected in either the same region or a separate region some distance downstream. The advantages of the systems are that they offer a method of measuring mass flow or linear velocity that is independent of velocity profile provided the range of the ionizing radiation is large compared with the dimensions of the pipe. Also, they are sensitive to rapid changes in flow and can be designed so that they operate with no, or very little, pressure loss. Applications occur at high and low flow rates, and, in principle, there is no serious limit to the cross-sectional area of pipe in which measurements can be made. With large-diameter pipes, however, a multielectrode structure is required, which will offer some impedance to the flow. Since an electric current is produced directly, this type of instrument is suitable for use in automatic control systems.

Ionizing Source and Detector at Same Location. In a system first described by Obermaier[28] (Fig. 7), annular electrodes comprising an ionization chamber are mounted in the gas stream and the gas in the space between them is ionized. An alpha emitter is used to provide high ionization density, and this is acceptable if the electrode separation is small. At low gas velocities most of the ions are collected by the applied field of the ionization chamber, but as the gas velocity is increased a higher proportion of ions is swept out of the chamber region. Thus the current decreases with an increase in gas velocity.

A quantitative relation between ionization current and gas velocity has been devised by Shteinbok,[29] who showed that, apart from gas velocity, the ionization current at a given gas temperature also depends on the humidity, pressure, and chemical composition of the gas.

The chief disadvantages of this method are the restricted operational range and the sensitivity of the collector current to collector voltage stability. In

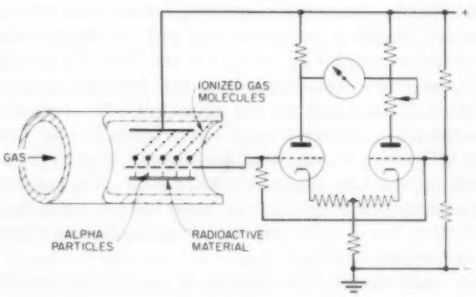

Fig. 7 Measurement of gas flow by induced-ionization system with ionizing source and detector at the same point.

common with other systems of gas-flow measurement of induced ionization, the collector is a high-impedance device, and therefore leakage currents in the electrode supports must be kept small (less than 10^{-12} amp). From this point of view the presence of water vapor and organic condensates in the gas can be troublesome and may invalidate general application.

Ionizing Source Upstream of Detector. The basic design of this system (Fig. 8) was first described by Blake.[27] At low flow rates the ions produced in the source region move downstream toward the ionization chamber but recombine before arrival. As the flow rate is increased, ions begin to arrive at the electrodes, and the collector current then continues to

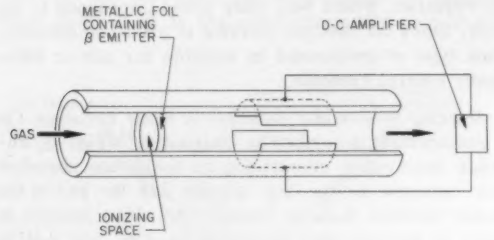

Fig. 8 Basic diagram of flow-measuring system with ionizing source upstream of the detector.

increase with increasing flow rate. If the variation in gas composition is not sufficient to affect the ionization current seriously, the instrument is capable of being calibrated to measure mass flow. An additional patent taken out by Mellen[30] also includes a means of determining flow direction. The distance between the source and the two ionization chambers (Fig. 9) is such that even when there is no flow a small ionization current is measured in each chamber. When the gas flows the instrument indicates both the direction and magnitude of the gas movement.

The operating range of the system shown in Fig. 8 depends on the source activity, the maximum ion transit distance available, and the measuring-equipment sensitivity. With a 6-mc $^{90}Sr-^{90}Y$ source, 7.5 cm from the electrodes, the working range is from 0.03 to 1 g of air per second at STP. This corresponds to a current variation between 5×10^{-10} and 6×10^{-9} amp. It has been suggested[31] that the upper limit of velocity measurement, but not of mass flow, may be increased by slowing down the gas velocity by mounting the source and detector in a length of pipe of increased diameter.

An analysis of this system of gas-flow measurement was made[31] recently to determine, both theoretically and experimentally, how such factors as the strength of the ionizing source and the separation

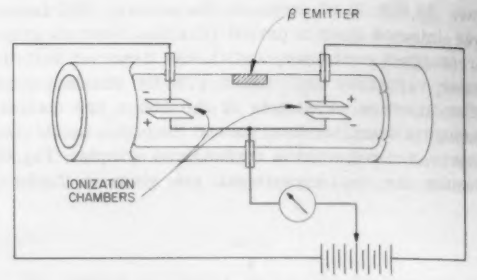

Fig. 9 Basic system to determine direction and velocity of gas flow. The two ionization chambers are sufficiently close to the source for a small current to be detectable when there is no flow.

of source and detector affect the operating range of the instruments. Calculated curves showing the variation of collector current with mass flow in a 2.5-cm-diameter pipe are shown in Fig. 10 for selected values of ion transit distance. The initial part of each curve shows a predicted square-law relation, and there is also a useful quasi-linear region where the ion transit distance is less than 1000 cm. Experimental ion transit distances of 7.5 and 28.5 cm obtained with the instrument shown diagrammatically

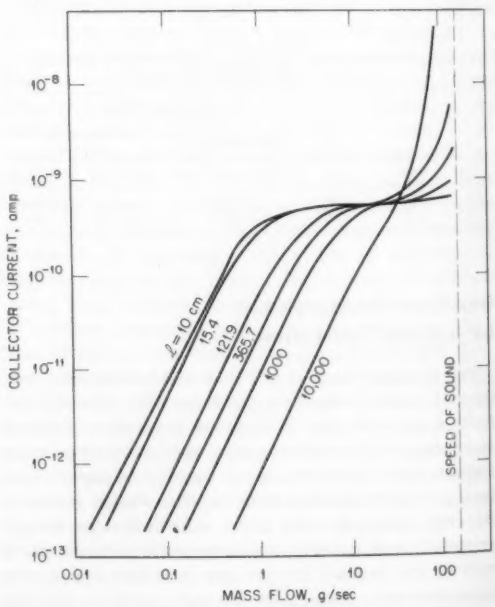

Fig. 10 Calculated curves showing variation of collector current with mass flow for several values of ion transit distance.

in Fig. 11 are given in Fig. 12. The agreement with calculated curves (Fig. 12) indicates the potential use of the theory for instrument-design studies.

The effect of gas impurities that change the values of ionization cross section and recombination coefficient is to change the number of ions entering the ion chamber and hence the measured current even though the mass flow is constant. Not all impurities are troublesome,[32] however (Fig. 13).

MEAN VELOCITY

In principle this method is the same as the isotope-velocity method, but in this case the ion transit time between two points of known separation is measured. In the original suggestion[33] a spark gap was used as the ion source, but in many applications this arrangement has the disadvantage that the presence of the electrodes introduces disturbances to the flow pattern. This is especially important at supersonic velocities. Radioisotope sources are intrinsically safe with regard to fire hazard compared to spark discharges, and this is important if the flow of flammable gases is to be measured.

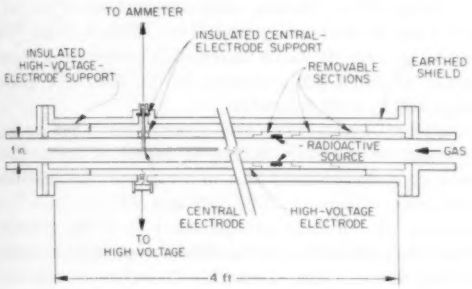

Fig. 11 Construction of flowmeter unit.

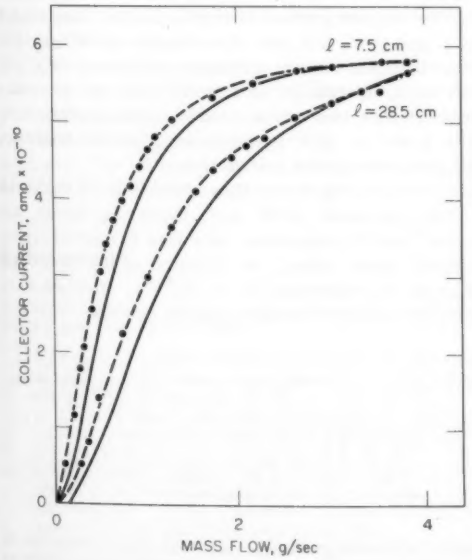

Fig. 12 Theoretical (—) and experimental (---) curves obtained with the instrument shown in Fig. 11.

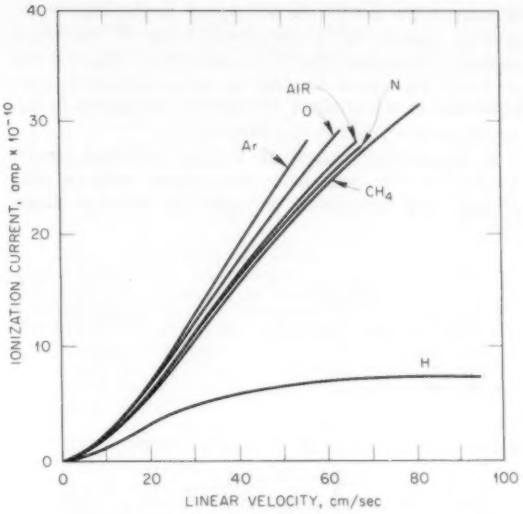

Fig. 13 The effect of using different gases in the flowmeter of Fig. 11.

When radioisotope sources are used, the ions are produced in bursts, either by interrupting a beam of alpha or beta particles with a high-speed mechanical shutter or by deflecting the particles with a periodically applied magnetic field.

It is essential that (1) the duration of each period of ionization be short compared with the transit time, (2) the gas velocity be large compared with the diffusion velocity so that the character of the ion pulse is not lost, (3) the ion collection time be short compared to the ion transit time, and (4) the phase shift in the amplifier be short compared to the ion transit time.

The main advantage of this method is that it is insensitive to impurities, and its principal limitation is that it measures linear velocity and not mass flow. However, a mass-flow value can be obtained if the gas density is measured simultaneously. A combined instrument of this type has recently been developed by Flanagan and Noe.[34]

Various methods of displaying available information have been suggested. Shumilovskii and Mel'ttser[35,36] suggested a circuit to measure the time delay directly, but a more useful technique is to use the current increase from the arrival of the ion wave

to trigger the modulator and thence to display the resultant signal as a recurrent train of electrical pulses. Since gas velocity is inversely proportional to ion transit time and this in turn is inversely proportional to recurrence frequency, the latter is directly proportional to gas velocity.

In the system[34] shown in Fig. 14, radiation from a 5-mc $^{90}Sr-^{90}Y$ source is interrupted with an aluminum disk containing an aperture through which

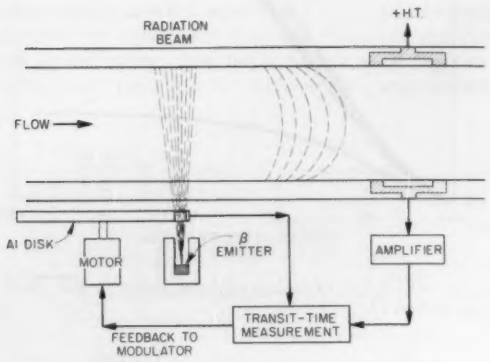

Fig. 14 Mean-velocity method of gas-flow measurement.

radiation passes at each rotation of the disk. An electrical signal synchronized with the ion wave is concurrently derived from a second aperture in the aluminum disk by using a small electric light bulb and phototransistor. This signal is arranged to trigger a multivibrator, which is then returned to its original quiescent state by a second signal derived from the arrival of the ion wave at the detector. The multivibrator output pulse length is thus directly proportional to the ion transit time.

Successive pulses from the multivibrator are smoothed to provide an output voltage proportional to transit time, t, and pulse frequency, f. The speed of rotation of the disk is automatically controlled to make this output voltage equal to a reference voltage, E. The pulse frequency is thus made inversely proportional to the transit time and directly proportional to the velocity, v. Hence $f = KE/t = KEv$. By further making the reference voltage proportional to gas density, the instrument can be made to measure mass flow. (This can be done by deriving E from the output of a radiation density gage.)

The practical range of these instruments depends on the ion transit distance, which in turn depends on transit time; this should always be less than 1 to 1.5 sec because of ion recombination. For a 3-m transit distance the upper limit of gas velocity exceeds 30 m/sec, while the lower limit is about 2 m/sec.

Accuracy depends on the alignment of source and collimators and on the stability of d-c levels in the trigger circuits. In practice these can be made less than 1%. With the disk system reproducibility is within 3%.

Other Uses of Radioisotopes in Flow Measurement

Radioisotope sources have been incorporated into rotating-vane, deflecting-strip, and hinged-gate type flowmeters, with a significant improvement in performance. Application to flow measurement of corrosive fluids at high temperature and pressure is particularly important since connections through walls can be avoided.

Shumilovskii, Gushchin, and Tolokonnikov[37] studied the design of a rotating-vane instrument of the turbine type to measure liquid flow. With a 1-mc ^{60}Co source on one or more vanes, this instrument is reported to have an accuracy better than ±1% in the range 300 to 1100 liters/hr.

Carlson, Cederberg, and Ljunggren[38] used an instrument of similar design to measure water flow rates in a downcomer pipe in a high-pressure boiler containing water at 2000 psi and 635°F. Two ^{60}Co sources, one of 3 mc and the other of 10 mc activity, were incorporated into two vanes separated by 120°. The use of two sources enables flow direction as well as velocity to be measured. With this arrangement the measured circulation started about 10 min after the first burner was ignited. Before steady state was reached, the water velocity changed direction continually; at low pressures highly damped oscillations were observed, but they disappeared at 140 psi. Results obtained during extraction of steam (Fig. 15) indicate an instability in flow velocity as the mainstream temperature is increased. Several other tests were made in this investigation, which illustrates well the value of this instrument.

For measuring water flow rates (0 to 70 cm^3/min) at high pressure (2000 psi), Cameron, Berry, and Taylor[39] used a rotameter operating in high-pressure stainless-steel tubing, the position of the float being observed by detecting 80 μc of ^{60}Co, attached to it, with a scintillation counter outside the pipe.

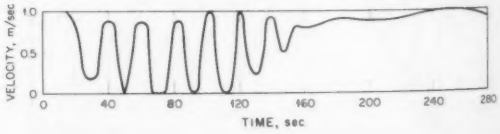

Fig. 15 Circulation instability during steam extraction as the temperature of the main stream is increased. Observations obtained with a rotating-vane flowmeter incorporating a radioactive isotope.

Flow measurement based on deflection of a thin elastic lamina was discussed by Shumilovskii and Mel'ttser.[40] The deflection in this case is observed by attaching a radioactive source to the moving element.

Clayton[41] suggested a hinged gate for low-velocity (1 to 1000 cu ft/hr) gas-flow measurement. One design (Fig. 16) has the source mounted at the edge of

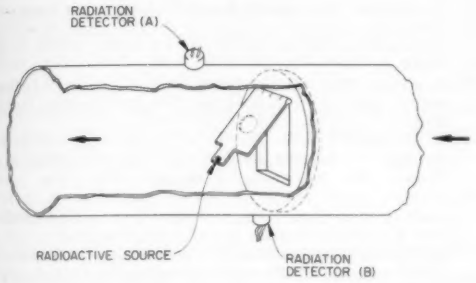

Fig. 16 Hinged-gate method of flow measurement: detector position (A) for gas-flow measurement, (B) for controlling liquid flow.

the gate and the deflection observed with a radiation detector (position *A*) outside the pipe. A wide operating range is possible, and it can be widened by using several gates in the same pipe section or even on the same hinge, each gate subtending different areas to the flow. If a detector with a suitable current–dose-rate characteristic is used, the operating characteristic is almost linear.

If the detector is mounted in position *B*, it can be used to control the flow rate of liquids. Because of the inverse-square law, exponential absorption of radiation in the fluid and the close approach of the source to the detector in the zero position, a high sensitivity to flow velocity is obtained. (MG)

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Radioisotopic Life Detector for Space Studies*

Abstract: The Gulliver space probe will use radioisotopes—^{14}C and ^{35}S—in the form of labeled components of a nutrient broth to demonstrate the presence of living organisms on Mars. The technique that has been developed was selected because of its sensitivity and the rapidity with which it can be carried out.

Gulliver, the National Aeronautics and Space Administration's biochemical probe named for Jonathan Swift's famous traveler to strange places, is designed to demonstrate, with the use of ^{14}C and ^{35}S, the presence of living organisms on Mars by detecting metabolism, growth, and photosynthesis. The radioisotope technique is characterized by sensitivity and rapidity—important features for an experiment that must be performed in a hostile environment on possibly unknown organisms of unknown metabolic rate.

In designing a life-detection system, it was assumed that, if life exists on other planets, microorganisms are an important part of the biosphere there as they are on Earth. Also, chemical considerations mark carbon rather than silicon, for example, as the most likely element forming the frame of organic compounds. Furthermore, the Martian atmosphere is known to contain large amounts of carbon dioxide. Metabolism and growth were selected as the basic processes to be detected even though our incomplete knowledge of Martian conditions and organisms makes design of specific metabolic experiments very difficult and uncertain. However, these continuous processes would offer the most conclusive evidence for life. In 1963 the Gulliver experiment was modified to include a photosynthesis detector, since many biologists believe that, if life exists on Mars, it must include photosynthesis organisms and may, in fact, even be limited to these.

Two other candidates for the life-detection job on Mariner B, scheduled for launch in 1968, are the Multivator, designed to detect fluorescence resulting from an enzymatic reaction taking place in microorganisms; and the Vidicon microscope, designed to seek visible signs of life with a lens system able to cover a 100-μ object field with 0.5 μ resolution and to relay back pictures in at least four shades of gray.

Metabolism and Growth

Based on terrestrial experience, it is commonly considered that the metabolic products of all organisms include gases. Gulliver therefore provides for release of the gases $^{14}CO_2$ or $H_2{}^{35}S$ from labeled

*The project scientists for the National Aeronautics and Space Administration are Drs. Gilbert V. Levin of Hazleton Laboratories, Inc., and Norman H. Horowitz of the California Institute of Technology. Dr. Levin kindly reviewed this summary, which is based on the references listed at the end.

substrates by microorganisms and measurement of the amount of these gases released. Since the rate of CO_2 production increases exponentially in growing cultures, the rate at which labeled CO_2 is released will indicate whether or not growth is taking place. Data obtained will be telemetered back to Earth.

The essential components of the probe (Figs. 1 and 2) are (1) a sampler for collecting microorganisms; (2) a nutrient broth containing radioisotope-labeled organic compounds; (3) an incubation chamber; and (4) a radioactivity detector for examining the metabolic products. In numerous terrestrial runs the metabolism-and-growth operation was successfully demonstrated, and results were obtained in minutes or hours (Fig. 3), in contrast to classical bacteriological testing methods, which require a day or so.

SAMPLER

The mechanism for collecting a sample of microorganisms consists of two 25-ft lengths of string, which are coated with silicone grease to make them sticky. These strings will be fired from inside the capsule and deployed over the surface of the planet. After a short time they will be reeled back, together with adhering soil particles, into the incubation chamber. Here any organisms present can use a nutrient broth for metabolism and growth.

NUTRIENT BROTH

Mediums that will evoke significant responses from all test organisms present in widely varying types of soil and in numerous types of pure cultures have been developed from two different starting materials:

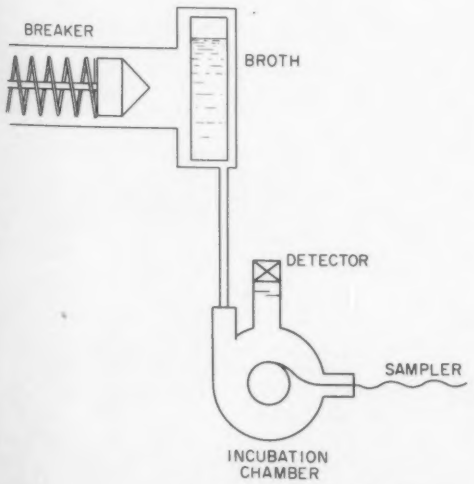
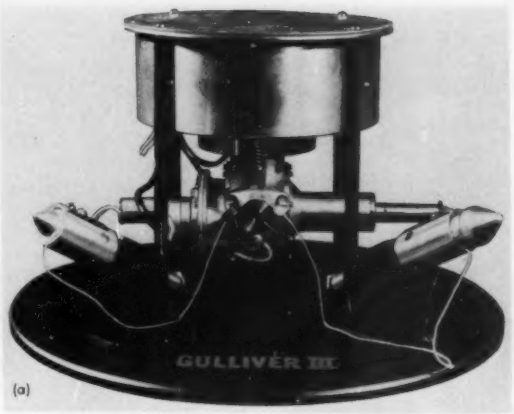


Fig. 1 Principle of the Gulliver radioisotopic probe.



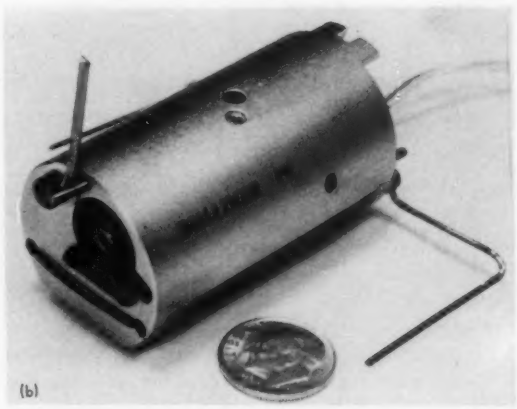


Fig. 2 Gulliver probe. (a) Model III (weighing 7 lb), showing projectiles that will deploy sticky strings on the surface of Mars for collecting samples. (b) Model IV, for in situ use, developed during the past year.

(1) a complex medium that was progressively changed as tested against numerous types of organisms and (2) a simple inorganic medium to which various substances were added, as needed, to elicit responses from the same organism. Many substrates have been tested, including

Sodium formate-^{14}C	Cysteine-^{35}S	Yeast extract-^{14}C
Sodium acetate-1-^{14}C	Glycine-2-^{14}C	*Escherichia coli* extract-^{14}C
Sodium lactate-^{14}C	Glucose-^{14}C	

In extraterrestrial experiments both D and L forms of optically active components of the medium will be included since it is not known which form might be required by an alien organism.

Organisms that have been tested include bacteria, streptomycetes, fungi, and algae (aerobes, anaerobes, heterotrophs, phototrophs, autotrophs, thermophiles, psychrophiles, mesophiles, spore formers, etc.).

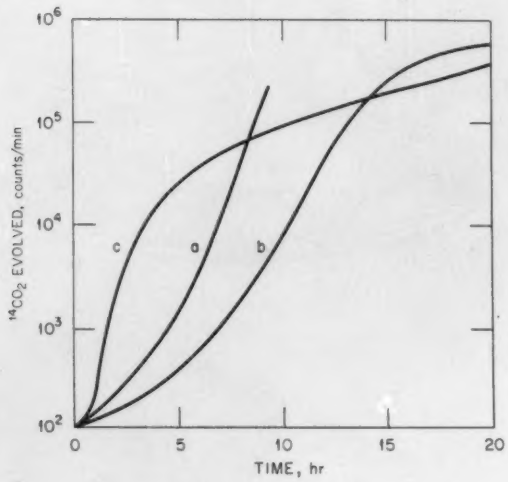


Fig. 3 Evolution, in terrestrial tests, of $^{14}CO_2$ from ^{14}C-labeled nutrient broth as a result of action of microorganisms found on (a) field soil, (b) asphalt, and (c) desert soil.

INCUBATOR

When the sampler line to which Martian soil is adhering has entered the incubation chamber, the ampule containing the broth will be broken, releasing broth onto the line. Any metabolically evolved gases will diffuse into the free space above the liquid and deposit on a thin layer of getter (barium or lithium hydroxide is used for carbon dioxide) on the radiation detector, which will measure its radioactivity. A baffle between the detector and broth will prevent the detector from "seeing" the broth directly but will not prevent the gas from reaching the getter.

RADIOACTIVITY DETECTOR

The beta-particle detector is a thin-window Geiger tube, which is the best detector for size, weight, power, and stability considerations. A ring of Geiger tubes surrounds the beta detector for anticoincidence purposes. Suitable preamplifiers, etc., are included.

Photosynthesis

Numerous field trials have demonstrated the workability of the photosynthesis detector. In the initial design ^{14}C-labeled carbohydrates were offered to photosynthetic organisms, and $^{14}CO_2$ evolution was monitored during alternating exposures of the organism to light and dark. In a subsequent simplification of the system, the organism is exposed to $^{14}CO_2$ in light to permit fixation of the gas. Unassimilated $^{14}CO_2$ is then exhausted from the chamber, and the organism is maintained in darkness while $^{14}CO_2$ evolution is monitored. Any CO_2 evolved here must come from intracellular products photosynthesized during the initial exposure to light. Attempts will also be made to measure the amount of labeled carbon remaining in the system. (Martha Gerrard)

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Synthesis of Radioisotope-Labeled Organic Compounds*

By P. Ph. H. L. Ottot

Abstract: Methods used for synthesizing labeled organic compounds are reviewed. Standard organic-chemistry syntheses are used whenever feasible. Special methods include the Wilzbach method, in which tritium is exchanged for hydrogen under the influence of the tritium radiation, and neutron irradiation of ferrocene to convert the ^{58}Fe in the molecule to ^{59}Fe. Some labeled compounds are formed by biosynthesis from a labeled substrate. The labeled compounds are purified by various methods, and the purity is determined before use, particularly if the labeled material has been stored.

The demand is continually increasing for labeled organic compounds, which may be supplied commercially or prepared as one step of a tracer experiment. This review discusses the basis for selection of the labeled material that is to be used, preparation methods, purification of the products, product purity determination, and storage.

A compound is labeled when one of its elements has an isotopic composition different from that in nature. For example, in an ordinary carbon compound, the carbon is practically all ^{12}C. If we replace part of the ^{12}C by ^{14}C, the compound is radioactively labeled. The degree of replacement is indicated by the specific activity, the maximum value being reached when all the carbon is ^{14}C.

The majority of organic compounds are made up chiefly of carbon, hydrogen, oxygen, nitrogen, phosphorus, sulfur, the halogens, and a few metals. Radioisotopes of only very short half-lives are known for oxygen and nitrogen, and only one radioisotope of satisfactorily long half-life is known for the others. Only about 10 of the some 1250 known radioactive nuclides are generally used for synthesizing labeled organic compounds. The most important ones, shown in Table 1, are all pure beta emitters except ^{131}I. The shortest lived radioisotope that has been introduced into an organic molecule is ^{11}C, with a half-life of 20.5 min, which was used 20 years ago when ^{14}C was not available and recently has been used again.

There are considerable differences in the maximum specific activity obtainable with these radioisotopes. The theoretical maximum is obtained only with tritium. The most limited is ^{36}Cl, because of its long half-life and the fact that ^{36}Cl made by a (n,γ) reaction on ^{35}Cl is usually diluted with inactive carrier. Compounds of ^{14}C having a high specific activity are expensive because of the isotope cost.

Table 1 RADIOISOTOPES USED MOST WIDELY FOR LABELING ORGANIC COMPOUNDS

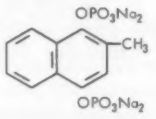
Isotope*	Radiation		Highest obtainable specific activity	Specific activity of labeled compounds, mc/mmole
	$E_{max.}$, Mev	Half-life		
^{3}H	β 0.018	12.3 years	60 curies/mmole H_2, >99% ^{3}H	10 to 10^{3}
^{14}C	β 0.155	5568 years	25 mc/mmole $BaCO_3$, 40% ^{14}C	1 to 10
^{35}S	β 0.167	87 days	>30 curies/mmole H_2SO_4, >2% ^{35}S	1 to 10
^{32}P	β 1.71	14.5 days	>160 curies/mmole H_3PO_4, >2% ^{32}P	1 to 10
^{36}Cl	β 0.71	3.1×10^{5} years	7.2 mc/mmole HCl, 0.7% ^{36}Cl	
^{131}I	β 0.61 γ 0.36	8 days	>3.3×10^{3} curies/mmole NaI, >20% ^{131}I	1 to 500

*Iodine-125, with a half-life of 56 days and a 0.036-Mev gamma emission, is now available.

Choice of Labeled Compound

CHOICE OF RADIOISOTOPE

The isotope used is selected partly on the basis of its special properties. For example, in autoradiography of tissues, the soft beta radiation of tritium is advantageous because it makes photographic images with very good resolution of detail. In brain-tumor diagnosis the penetrating gamma radiation of ^{131}I-labeled diiodofluorescein is advantageous because it can be detected by instruments located outside the skull. And in the treatment[1] of malignant tissues, the compound Synkavit,



which is taken up by malignant tissue in much higher concentration than by normal tissue, is labeled with

*Abridged from *Chemisch Weekblad*, 60: 59-68 (1964) (in Dutch; English translation by Martha Gerrard, ORNL-tr-1200).

†TNO Central Laboratory, Delft, Netherlands.

tritium and used to carry a therapeutic amount of radioactivity to the tumor. The low-energy beta emission of tritium affects only the cells where the drug localizes. The high specific activity required could also be obtained with ^{32}P, but its higher energy beta radiation would affect normal cells also.

SPECIFIC ACTIVITY AND AMOUNT

The specific activity and the amount of a labeled compound needed for a tracer experiment depend largely on the nature of the experiment and the manner in which it is to be carried out. The scale of the experiment, the chemical form of the material on which measurements will be made, the radiation detector, and the accuracy required for the measurements must all be considered.

Too high a specific-activity requirement and too large an amount of compound may make the synthesis and the tracer experiment unnecessarily expensive. For example, in an actual experiment conducted on a 100-ml scale, the reaction products were separated chromatographically, and the yield was determined by weighing. Then the effects of various experimental conditions were determined with labeled compounds, and yields were measured by thin-layer chromatography and scanning instead of by the difficult and time-consuming preparative chromatographic separation. In a large-scale experiment, if the starting material is labeled with ^{14}C, a specific activity of 1 mc/mmole is required, and 10 mmoles of this material costing, for example, $70 per millicurie, would result in a total isotope cost of $700 per experiment. If the scale of the experiment is reduced by a factor of 100, to 1 ml, the isotope cost is only $7. And in scouting experiments, where great accuracy in the determination of product yield is not needed, thin-layer chromatography can be combined with autoradiography. Then the required specific activity is 10 μc/mmole, and the isotope cost is decreased to 7 cents per experiment.

LOCATION OF RADIOISOTOPE IN THE MOLECULE

In most cases, not all atoms in a molecule are labeled; the activity may be in one distinct location, at several distinct locations, or distributed over the whole molecule either uniformly or nonuniformly. It is important to know both the location of the label and its fate in the reaction in which the labeled compound is to take part. The radioactive atom must be bound in the molecule in such a way that the radioactive product measured at the end of the experiment is representative of the group in the molecule that we are interested in. Otherwise results will be misleading.

AVAILABILITY OF LABELED COMPOUNDS

Labeled compounds may be obtained from various sources. The IAEA distributes the International Directory of Radioisotopes,[2] which lists all commercially available compounds. *The Isotope Index*[3] is a similar listing and includes prices.

For some time there has been a Euratom collection of labeled compounds made in research laboratories in larger quantities than needed.* These compounds can be obtained from Euratom in Brussels (European Atomic Energy Community, Directorate-General, Research and Information on Radioisotopes, Belliard St. 51-53) for a moderate fee.

When a compound cannot be obtained commercially, it can sometimes be synthesized in a specially designed laboratory for the purpose. The Isotope Division of the TNO Central Laboratory (P. O. Box 217, Delft, Netherlands) has such a laboratory where labeled compounds can be prepared to order. In limited numbers, outsiders can be admitted as guests to make their own syntheses.

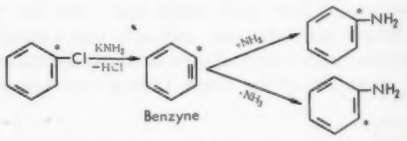
Preparation Methods

Labeled compounds are prepared chiefly by standard organic-chemistry syntheses,[4,5] but a few special methods are also used.

ORGANIC-CHEMISTRY SYNTHESES

Carbon-14, ^{35}S, and radioisotopes of other polyvalent elements that form the molecular skeleton of organic compounds are usually introduced into the molecule by reactions in which the skeleton is synthesized. For complex molecules, however, this can be a laborious and costly undertaking. Tritium, the halogens, and other univalent elements, which are found at the periphery of the molecule, often may be simply added to an unsaturated compound or exchanged with a stable isotope already in the molecule. In general, we try to introduce the radioisotope in the last step of the synthesis in order to hold the number of manipulations with radioactive material to a minimum and thereby achieve a maximum yield of the radioactive product.

Additional requirements may be to place the label in a specific location and to preferentially form one isomer of a compound that exists in two isomeric forms. Sometimes intermediates give rise to a distribution of the label over several positions. For example, 1-chlorobenzene-1-^{14}C reacts with potassium amide in liquid ammonia to form[6] two anilines, one labeled in the 1 position and the other in the 2 position:



*See *Isotopes and Radiation Technology*, 3(3): 263-267 (Spring 1966).

Similarly, in the reaction of allyl alcohol-1-^{14}C with thionyl chloride, the product obtained is labeled in the 1 or 3 position, or both, depending on the reaction conditions.[7] Propene-^{14}C may be prepared[8] by dehydration of propanol-1-^{14}C in the presence of heated aluminum chloride or metaphosphoric acid to give an equimolar mixture of 1- and 3-^{14}C-propene-1:

$$CH_3CH_2{}^{14}CH_2OH \xrightarrow{-H_2O} \begin{matrix} CH_3CH={}^{14}CH_2 \\ {}^{14}CH_3CH=CH_2 \end{matrix}$$

However, pure 1-^{14}C-propene-1 can be prepared by the Hofmann reaction after conversion of propanol-1-^{14}C to the bromide with phosphorus and bromine and formation of the quaternary ammonium base with trimethylamine and silver oxide:

$$CH_3CH_2{}^{14}CH_2OH \xrightarrow{P,Br_2} CH_3CH_2{}^{14}CH_2Br$$
$$\rightarrow CH_3CH_2{}^{14}CH_2N(CH_3)_3OH \rightarrow CH_3CH={}^{14}CH_2$$

Specifically labeled pentenes are synthesized[9] by the reaction of allyl magnesium chloride and ethyl-1-^{14}C or ethyl-2-^{14}C iodide:

$$CH_2{=}CHCH_2MgCl + CH_3{}^{14}CH_2I \rightarrow CH_2{=}CHCH_2{}^{14}CH_2CH_3 + MgICl$$

$$CH_2{=}CHCH_2MgCl + {}^{14}CH_3CH_2I \rightarrow CH_2{=}CHCH_2CH_2{}^{14}CH_3 + MgICl$$

To demonstrate the specificity of labeling, the molecule must be degraded.

Labeled compounds of high specific activity are usually made in small amounts by semimicro- or micromethods. For working with small volumes of volatile compounds of high specific activity, special high-vacuum techniques have been developed. For example, small volumes of volatile compounds are condensed with liquid air in a high vacuum and quantitatively transferred from one container to the other in apparatus like that shown in Fig. 1. In order to facilitate diffusion, wide connections are used; the vacuum is created by a high-speed diffusion pump. In reactions using tritium gas as a tritiating agent, since tritium cannot be condensed with liquid air, it is transferred with a Toepler pump. The connections here are made as small as possible.

Synthesis with radioisotopes requires considerable preliminary work, and the reactions are first carried out repeatedly with nonradioactive material. Radioactive syntheses can, in practice, be surveyed by means of a well counter, with which the radioactivity of the solution is measured. The method can be easily standardized and is used in determining the radiochemical yield and specific activity of compounds labeled with ^{14}C, ^{35}S, ^{32}P, ^{131}I, ^{36}Cl, etc. The sensitivity for ^{14}C and ^{35}S is not great, the lowest limit being about 1 μc/ml. However, this is sufficient for high-specific-activity material. An advantage of using this instrument is that the wanted material is not diluted with inactive material. After the measurements have been made, the compound can be returned to the process so that essentially no labeled product is lost during isotope assay.

SYNTHESIS USING NUCLEAR RADIATION

In tritiation by the Wilzbach[10] method, the chemically pure compound is exposed to high-purity tritium gas, whose ionizing radiation causes it to exchange with hydrogen. A difficult synthesis can thus be avoided. However, radioactive by-products of high specific activity are formed and are usually difficult to remove. Therefore we consider this method a last resort for labeling compounds that are especially difficult to obtain by chemical synthesis (e.g., proteins and nucleic acids). Tritiation is also useful with unsaturated compounds, which are hydrogenated in good yield, and with iodine-containing compounds, in which the iodine is preferentially replaced by tritium.

Irradiation of organic compounds with reactor thermal neutrons is a special method that has been used successfully, for example in the irradiation of crystalline ferrocene[11] to convert the ^{58}Fe to ^{59}Fe. A neutron enters the target nucleus, and a number of gamma rays are emitted. Through the resulting recoil the iron atom obtains a kinetic energy of several hundred electron volts, and the iron-cyclopentadiene bonds are ruptured (chemical binding energies are generally less than 5 ev). Under the conditions of the experiment, however, ^{59}Fe-ferrocene is re-formed in 20% of theoretical yield, which is particularly high. In most other cases studied, however, yields have usually been less than 1%, and the activity has been found almost entirely in the unwanted and difficult-to-remove decomposition products.

BIOSYNTHESIS

Labeled compounds have long been made by biosynthesis.[12] Glucose and other carbohydrates uniformly labeled with ^{14}C are formed in satisfactory yield in tobacco leaves by photosynthesis from $^{14}CO_2$. Fatty and amino acids labeled with ^{14}C can be made by algae (*Chlorella vulgaris*). More recently, ^{75}Se-selenomethionine and ^{75}Se-selenocystine of high specific activity have been made by yeast cells.[13] Because of the great similarity in chemical and biological properties, ^{75}Se-selenomethionine is used as a tracer for methionine in organ scanning, the ^{75}Se being more easily detected externally than ^{35}S-labeled methionine because of its gamma emission.[14]

Much research remains to be done before such biologically interesting compounds as labeled vita-

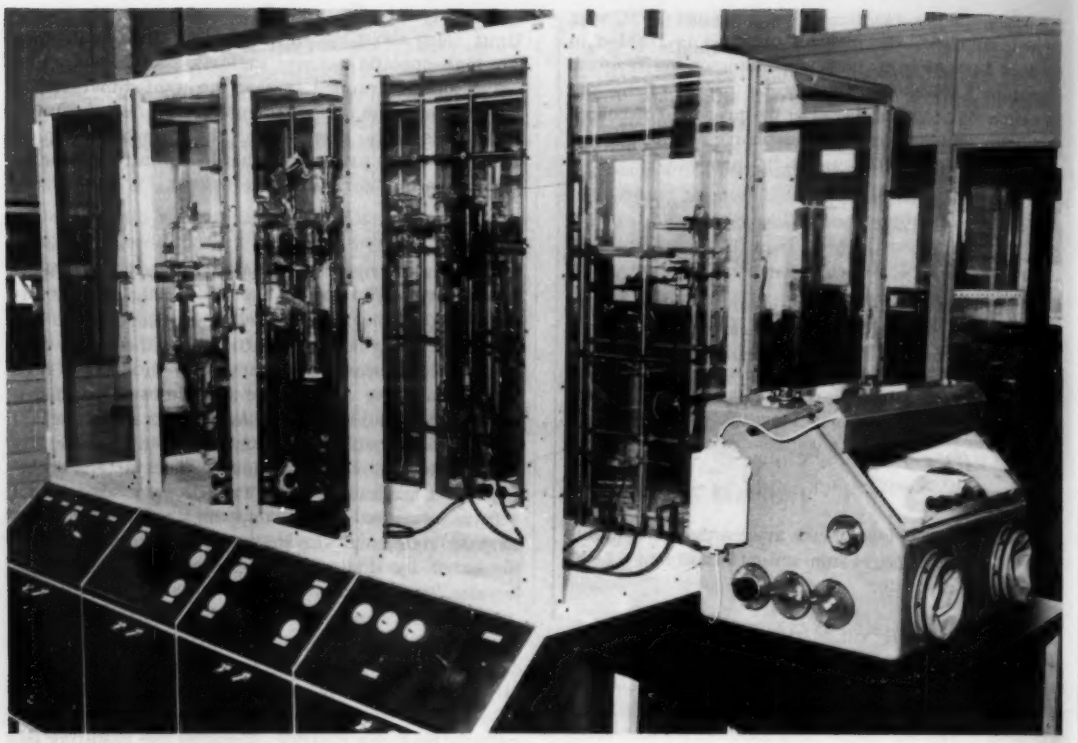


Fig. 1 High-vacuum apparatus for synthesis of labeled compounds. The apparatus is inside a closed hood from which the air is continuously drawn off to create a slight vacuum. There is a special service panel outside.

mins, hormones, and alkaloids, whose chemical synthesis is difficult, can be satisfactorily obtained by biosynthesis. Very low radiochemical yields and radiochemically impure products are typical of such preparations.

Purity

Both chemical and radiochemical purity are important in a labeled compound, but only the radiochemical purity is of concern in this paper. The radiochemical purity indicates what fraction of the radioactivity of a sample is that of the desired compound. A significant part of the radioactivity may be present in trace impurities, for instance, when inactive (carrier) material has been added, since radioactive by-products and contaminants are not diluted with the carrier. Because radioactive impurities can affect the results of a tracer experiment, the radiochemical purity of the material used must be determined.

For determining radiochemical purity, isotope dilution analysis can be used. Chromatographic methods combined with radiation detection are also useful, e.g., paper or thin-layer chromatography followed by autoradiography or scanning; column chromatography combined with scintillation counting; and gas chromatography combined with measurement of radioactivity in the effluent by means of an ionization chamber. An automatic instrument (Fig. 2) is useful for scanning the thin-layer and paper chromatograms. A large variety of equipment is now available commercially. In Fig. 3 are shown the results of a determination of the purity of a compound of low specific activity by combination of thin-layer chromatography and autoradiography.

PURIFICATION

For purification of labeled compounds besides the classical methods, preparative chromatographic methods are used: column chromatography, preparative gas chromatography, thin-layer chromatography, and thick-paper chromatography. These chromatographic methods combine very well with syntheses carried out on a micro- or semimicroscale. In

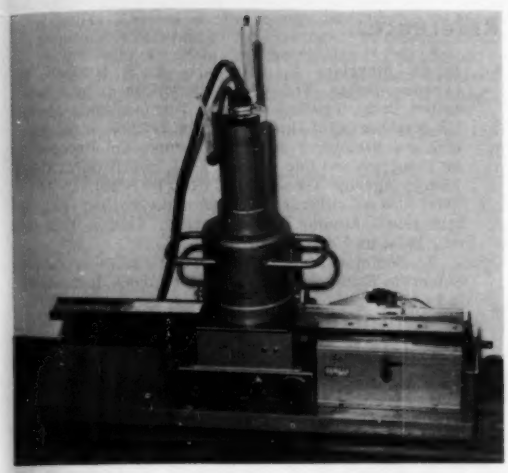


Fig. 2 Apparatus for automatic scanning of a thin-layer chromatogram. The glass strip with the chromatogram is fastened to the slide and drawn slowly under the detector. The movement may be at a constant rate or in steps of 1 to 5 mm. A rate meter and a recorder are used in the continuous scanning, and a scaler and a printer are used in the step scanning.

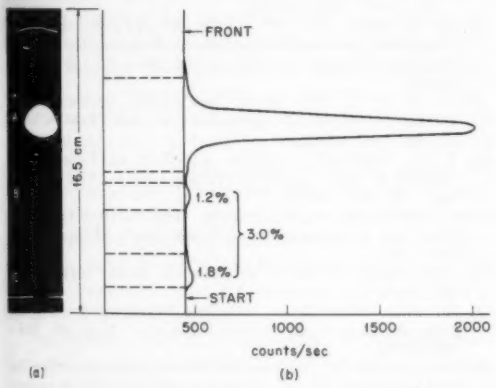


Fig. 3 Autoradiographic determination of purity of phenyldioxane-^{14}C of low specific activity (10 mμc/mg). Besides the principal peak in A, two small peaks of contaminants can be seen. The curve in B was made by scanning the film, with the apparatus shown in Fig. 2, after the silver had been quantitatively converted to $Ag^{131}I$.

Table 2 ANALYSIS OF 5-^{14}C-PENTENE-1

Component	Before purification		After purification	
	Amount, mole %	Radioactivity, %	Amount, mole %	Radioactivity, %
Ethane (ethene)	>1.2	>1		
Propane	>0.007	0.02		
Propene	>0.11			
Isobutane	<0.001	0.2		
n-Butane	0.02	0.4		
Butene	0.1	0.3		
Isobutane	0.3	0.3		
n-Pentane	0.001	0.04		0.3
Pentene-1	79	78	100	99.7
cis-2-Pentene	<0.001	0.05		
2-Methylbutene	<0.001	0.07		
Not known	<0.001	0.1		
Not known	<0.001	0.07		
n-Hexane?	<0.001	0.02		
1-Hexene	<0.001	0.14		
Iodoethane	20	19.3		

Table 2 are shown the chemical and radiochemical purities of a ^{14}C-pentene sample before and after purification by gas chromatography.[9] In this case the contamination probably came primarily from the ^{14}C-ethyl iodide, which was 2 years old when used and could have undergone some autoradiolysis. It was diluted with inactive ethyl iodide for the reaction.

STORAGE OF LABELED COMPOUNDS

During storage of labeled compounds, some of the radioactive atoms decay, the rate being 0.01% per year for ^{14}C, for example. Significant decomposition[15,16] of the material may be caused by the ionizing radiation emitted, which is usually absorbed. Any labeled decomposition products cause radiochemical contamination. The radiation dose received in a year by a ^{14}C-labeled compound with a specific activity of 1 mc/mmole is more than 10^7 rads (molecular weight 125). One labeled with ^3H and having a specific activity of 100 mc/mmole receives 10^8 rads. The decomposition caused depends largely on the radiation sensitivity of the compound and the storage conditions, e.g., temperature, presence of oxygen, and presence and nature of a solvent or inert diluent, which may absorb a large part of the radiation. In some cases the crystal form is important; e.g., choline chloride is especially sensitive to radiation at room temperature but above 80°C is converted to another crystalline form in which its radiation sensitivity is normal.[17,18]

Of the various classes of organic compounds, aromatics are the most resistant to radiation (Table 3),

Table 3 COEFFICIENT OF RADIATION DECOMPOSITION

Compound	Coefficient, molecules/100 ev	Compound	Coefficient, molecules/100 ev
Saturated hydrocarbons	6–9	Carboxylic acids	5
Unsaturated hydrocarbons	14–2000	Esters	4
Aromatic hydrocarbons	1	Organic halogen compounds	4
Alcohols	4–12	Quaternary ammonium salts	5–2000
Ethers	7	Amino acids	2–30
Ketones	7		

and aromatic solvents such as benzene even have a protective effect on dissolved substances. Unsaturated carbohydrates are very sensitive to radiation, as are ^{14}C-cholesterol and ^{14}C-methylcholine chloride (Table 4). On the other hand, the radiation resistance of cyclooctatetraene, an otherwise very reactive compound, is comparable to that of benzene.[19]

Table 4 DECOMPOSITION OF LABELED COMPOUNDS AS A RESULT OF SELF-ABSORPTION

(Calculated Radiation Dose: 10^7 Rads)

Labeled compound	Specific activity, μc/mg	Decomposition, %
Valine-4,4'-^{14}C · HCl	6.0	1–2
Norvaline-3-^{14}C · HCl	17.7	5
Norleucine-2-^{14}C	17.4	2
Methyl-^{14}C-choline · HCl	13.0	63
Calcium glycolate-1-^{14}C	5.8	13
Calcium glycolate-2-^{14}C	4.0	22
Cholesterol-4-^{14}C	6.5	~40
Glycine-2-^{14}C · HCl	4.9	<1
Guanine-4-^{14}C · HCl	2.6	<1
Succinic acid-2-^{14}C	3.9	<1

Experience has shown that radioisotope-labeled compounds are best stored in sealed ampules under nitrogen or in vacuum, at low temperature, and dissolved in a protective solvent or adsorbed on a stable material. If a labeled compound is stored for a long time, its radiochemical purity should be determined again before it is used in tracer experiments.

ISOTOPE DEPARTMENT
OF THE TNO CENTRAL LABORATORY

Important phases of the Isotope Department's work are synthesis of labeled compounds and research with radioactive tracers. Among the projects of interest here are:

Synthesis: Labeled polymers for studying the effect of molds, biologically interesting compounds (e.g., ^{35}S-methionine, 2,4-dichlorophenoxyacetic acid-1-^{14}C, and chloramphenicol-T with the tritium exclusively in the aromatic nucleus[20]), ^{14}C-labeled hydrocarbons,[9] and tritiated polyphenyls.[21]

Research: Hardening of phenol-formaldehyde resins using compounds labeled at specific positions [e.g., 4-phenyl-1,3-dioxane-(2-^{14}C) and -(6-^{14}C)]; migration in polymers of certain additives (softeners, lubricants, and antioxidants); degree of modification of substituted amylose ethers; kinetics of copolymerization reactions using labeled monomers and of exchange reactions on polyvinyl chloride; reaction mechanisms in the catalytic conversion of propadiene to propyne, using a tritium-labeled catalyst[22]; racemization during Wilzbach tritiation of amino acids[23] and peptides. (MG)

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*Not available in Isotopes Information Center.

The Nuclear Analysis Facility of Texas Nuclear Corporation

By C. V. Parker†

Abstract: The Texas Nuclear On-Line Neutron Activation Analysis Facility was set up for industry to use in making feasibility studies on process-control applications. Material is run on a conveyor under a neutron generator, and the prompt and delayed gamma radiations produced are analyzed on a multichannel analyzer as a series of conveyors carries the material to a storage hopper.

The Texas Nuclear On-Line Neutron Activation Analysis Facility was established in cooperation with the Division of Isotopes Development of the U. S. Atomic Energy Commission to enable industry to perform feasibility studies on process-control applications based on fast-neutron inelastic scattering and fast-neutron activation analysis measurement techniques. This facility is available for use by any company or industrial organization, but the work is performed by Texas Nuclear personnel. However, any interested company is urged to send its own personnel to actively enter into the feasibility study. The study is performed and a report written at no cost to the participating organization.

The facility contains a bulk-material handling system and a data-acquisition and -analysis system. The material handling system consists of a 25-ton-capacity storage hopper, a 22-ft-long 18-in.-wide belt conveyor, a 2.5-ft-long forced-feed horizontal screw conveyor, a 31-ft-long 9-in.-diameter vertical screw conveyor, and a 25-ft-long 9-in.-diameter screw conveyor that feeds into the top of the hopper.

In the operation of the material handling system (Fig. 1), material is fed at the desired rate from the storage hopper via a sliding gate valve and chute arrangement onto the end of the belt conveyor. This conveyor moves the material past a leveling bar and under a vertically mounted neutron generator, whose neutrons impinge on the moving material. The neutrons are scattered inelastically by some of the nuclei in the material, and these nuclei promptly emit characteristic gamma rays. Some of the other nuclei undergo (n,γ) reactions. For example, in coal the carbon and oxygen nuclei are excited, and the silicon and aluminum nuclei are activated. The prompt gamma rays emitted by the carbon and oxygen atoms in the moving coal are measured with a prompt-gamma-ray spectrometer at the point of irradiation, and the delayed activity from the silicon and aluminum is measured downstream before the material is dumped into the forced-feed screw conveyor.

The material is then transported by screw conveyors back into the hopper, making this a continuous system. The speed of the belt conveyor can be adjusted from 10 to about 50 ft/min, and the system has an hourly capacity of about 25 tons of material weighing about 50 lb/cu ft.

A small, simple liquid loop system[1] has been added, consisting of 10 ft of 0.25-in.-OD Tygon tubing wrapped around the target assembly of a conventional neutron generator with another 10 ft wrapped around a 3- by 3-in. NaI(Tl) spectrometer housed in a lead and paraffin shield located about 50 ft away. A small electric pump forces the liquid through the tubing at various speeds, depending on the specific gravity of the solution. A 5-gal container is used as the reservoir. The sensitivity for measuring oxygen and aluminum in flowing mediums is being investigated.

The data-acquisition and -analysis system consists of an automated 100-kv neutron generator, a neutron monitor, a prompt-gamma-ray spectrometer, a delayed-gamma-ray spectrometer, and a data-analysis unit. The neutron generator is positioned with the target assembly about 0.5 in. above the moving material (Figs. 2 and 3).

†Texas Nuclear Corporation, Austin, Tex.

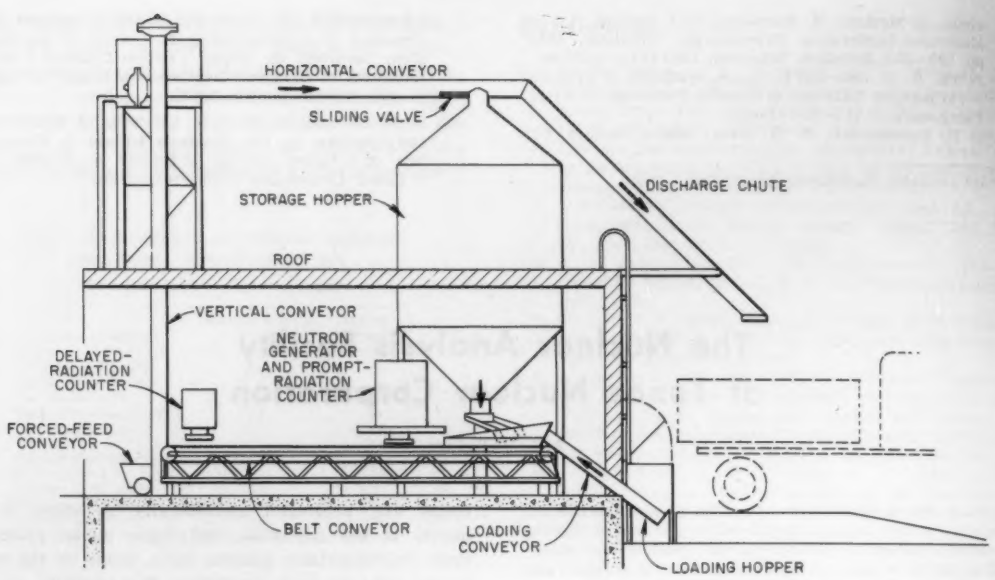

Fig. 1 Material handling system of the Texas Nuclear On-Line Neutron Activation Analysis Facility.

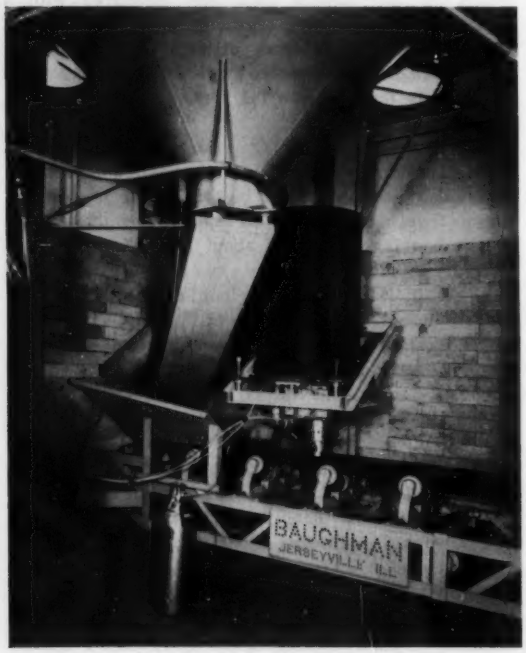

Fig. 2 The irradiation site, showing the neutron generator target, the bottom of the storage hopper, and portions of the prompt-gamma-ray spectrometer, conveyor belt, and screw conveyor.

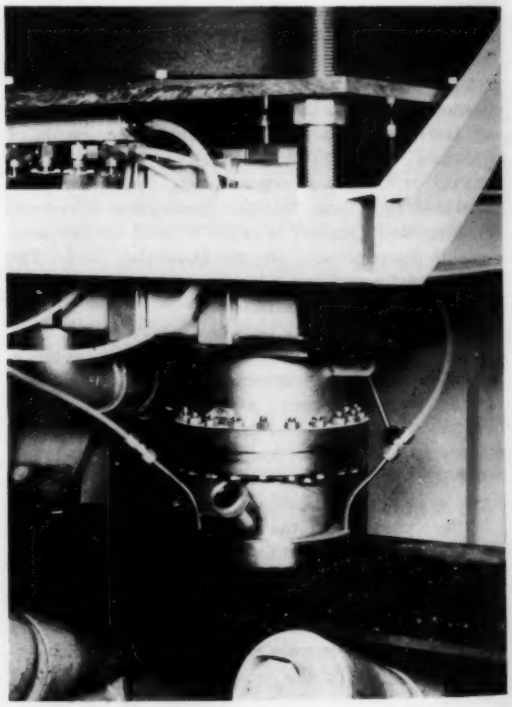

Fig. 3 A close-up of the irradiation site.

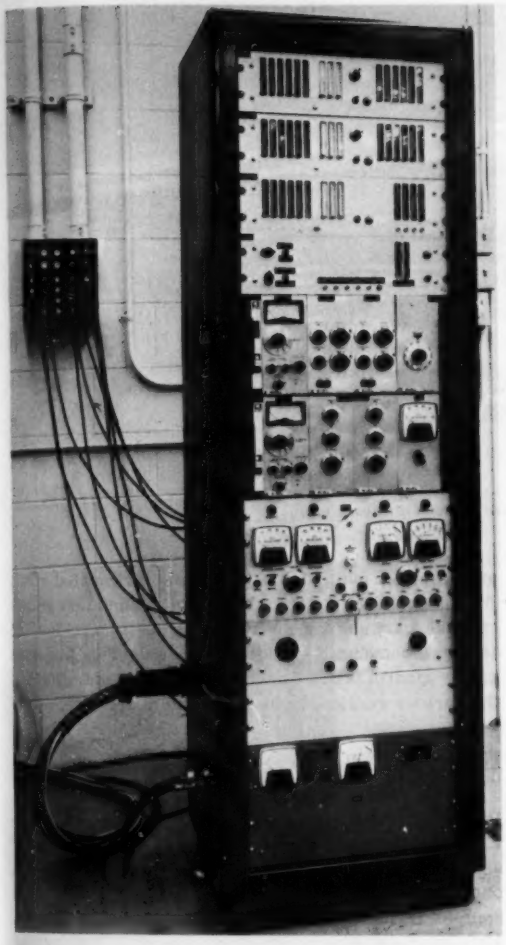

Fig. 4 Data-analysis system with the generator control panel and associated circuitry.

The analysis system (Fig. 4) can measure the yield of two elements each from the prompt and delayed gamma radiations emitted by the analyzed material. Figure 4 shows the data-analysis system with the generator control panel and associated circuitry. The counting yields from each element measured are recorded by scalers which correct for background radiation and Compton radiation. The neutron flux is also monitored and recorded on a scaler. At the end of a preselected counting interval, the outputs of the five scalers, the clock time, and the elapsed time are printed out, and the system is reset for the next count by the data-analysis control circuit. This circuit has manual as well as automatic control features.

The material handling system and the automated neutron generator and data-analysis unit have performed satisfactorily. The generator target has a lifetime greater than 75 hr at a neutron flux of approximately 2×10^9 n/(cm^2)(sec).

Additional information about the Texas Nuclear Facility can be obtained by writing or calling

Manager, Nuclear Analysis Facility
Texas Nuclear Corporation
P. O. Box 9267, Allandale Station
Austin, Texas 78756
Telephone: 512-452-8101

Feasibility studies can be scheduled by contacting

Mr. James W. Hitch
Isotopes Technology Development
Division of Isotopes Development
U. S. Atomic Energy Commission
Washington, D. C. 20545

Reference

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Activation Analysis at General Atomic

By H. R. Lukens, Jr.*

Abstract: The activation-analysis facilities at General Atomic Division, General Dynamics Corporation, San Diego, include 3 TRIGA reactors that can provide pulsed neutron fluxes up to 10^{17} n/(cm^2)(sec) as well as steady-state irradiators of lower flux levels; 2 General Atomic Linacs that can produce monoenergetic electron beams at any energy up to 45 Mev, and converters to produce bremsstrahlung for photoactivation analyses; 2 Texas Nuclear Cockcroft–Walton accelerators that can generate 2.5-ma beams of deuterium ions at 150 kv and can produce up to 2.5×10^{11} 14-Mev n/sec. Associated with this equipment are automated analytical systems that permit a wide spectrum of analyses by activation.

Activation analysis is a powerful method of elemental analysis based on the identification and quantitative measurement of specific radioisotopes subsequent to their generation during the irradiation (activation) of

*General Atomic Division, General Dynamics Corporation.

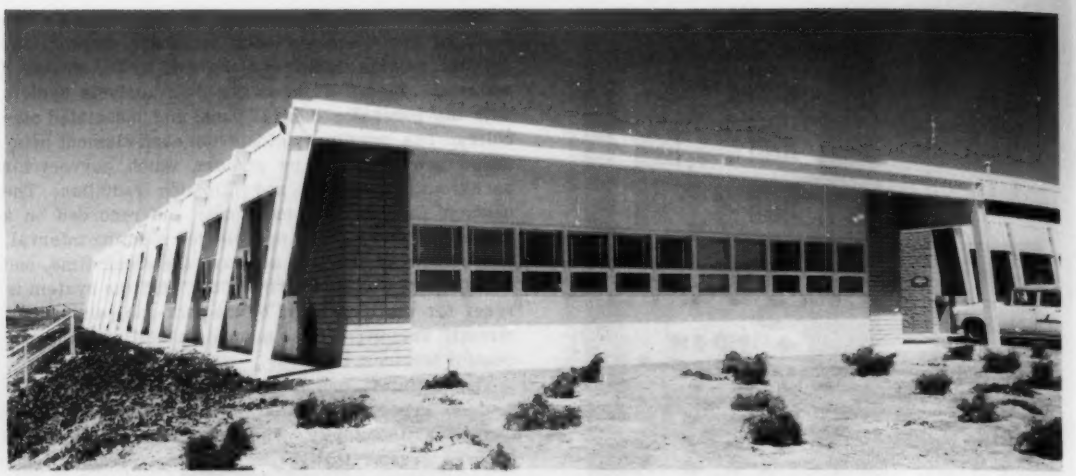

Fig. 1 General Atomic activation-analysis building.

a sample. A vigorous program of routine and research work in this area is among the many fields of research and engineering conducted by the General Atomic Division of General Dynamics Corporation. The facility, situated on a 350-acre site in the northern part of San Diego, Calif., is abundantly equipped with laboratories and hardware to permit a very full participation in the particular field of activation analysis.

The activation-analysis group is centralized in a single building devoted to this analytical method. The building (Fig. 1) is situated in a group of buildings consisting of a nuclear reactor facility (containing three pool type TRIGA reactors), an electron linear accelerator site (containing two Linacs), and a Cockcroft–Walton bunker (containing two C–W accelerators) and provides a wide variety of irradiation conditions. Included in the building are seven fully equipped radiochemical laboratories, a "cold" sample preparation laboratory, a gamma-ray-spectrometry and counting laboratory (shown in Fig. 2), a remote-control room for C–W experiments, a conference room, and offices.

Radioisotope Identification and Measurement

The gamma-ray-spectrometry and counting laboratory contains a number of multichannel pulse-height analyzers, single-channel analyzers, scalers, NaI(Tl) detectors, solid-state detectors, and gas-filled counters. A card-punch unit is connected to several of the multichannel gamma-ray spectrometers, which permits automatic data output to cards for computer processing. The card-punch unit is also connected to a multichannel gamma-ray spectrometer located at the TRIGA facility. Additional single-channel and multichannel gamma-ray-spectrometer units are located at the Linac and C–W sites. These devices are used in the identification and quantitative measurement of the various radioisotopes in activated samples.

IRRADIATION FACILITIES

TRIGA Reactors. The 250-kw (steady-state) TRIGA Mark I reactor (Figs. 3 and 4) provides fluxes of 4.3×10^{12} thermal n/(cm^2)(sec) and 3.5×10^{12} fission-spectrum n/(cm^2)(sec) at outer core positions. Several pneumatic transfer systems are used for in-core sample irradiations. A rotary specimen rack with 40 sample positions is generally used for lengthy

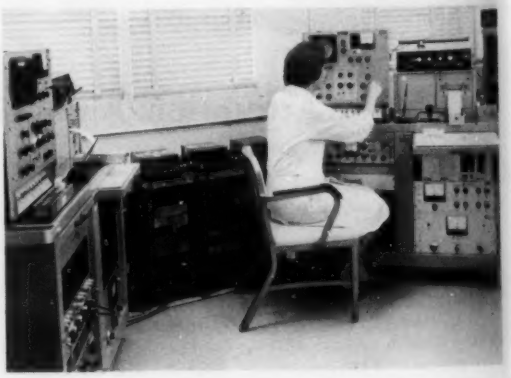

Fig. 2 Gamma-ray-spectrometry and counting laboratory.

irradiations. The rack rotates about the reactor core at 1 rpm, which ensures that all simultaneously irradiated samples receive the same dose. The thermal-neutron flux in this position is 1.8×10^{12} n/(cm^2)(sec) (at 250 kw).

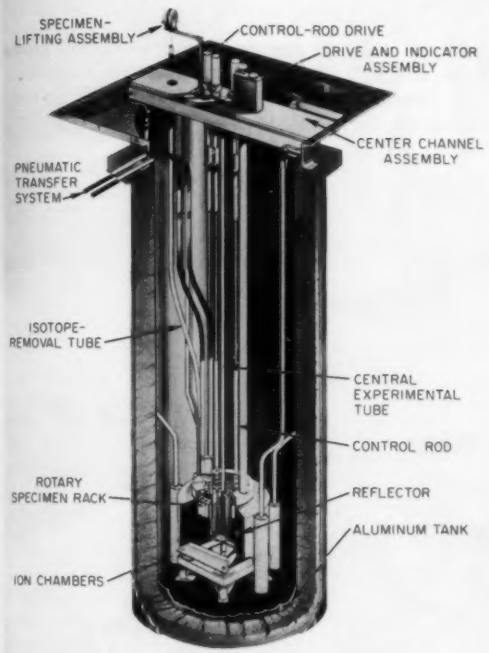

Fig. 3 TRIGA Mark I reactor cutaway.

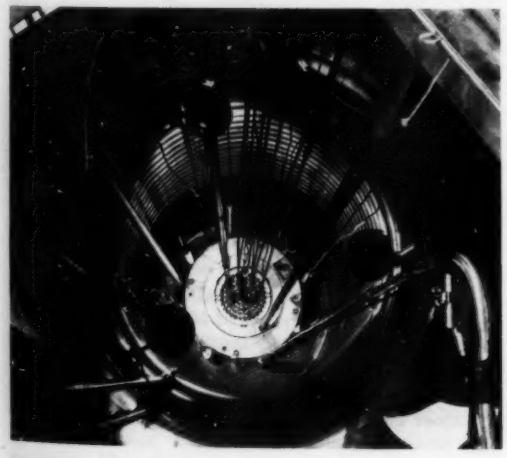

Fig. 4 TRIGA Mark I reactor installation.

A room adjacent to the Mark I reactor houses an advanced TRIGA prototype reactor (ATPR), which is capable of 1-Mw steady-state operation. This reactor is mounted on overhead rails so that it can be moved in the pool; this facilitates large irradiation experiments.

The third reactor is a newly installed 1-Mw (steady-state) TRIGA Mark III reactor, mounted on rails and equipped with a rotary specimen rack and pneumatic sample-transfer systems. This reactor (Fig. 5) is used primarily for steady-state irradiations of extended duration (weeks to months).

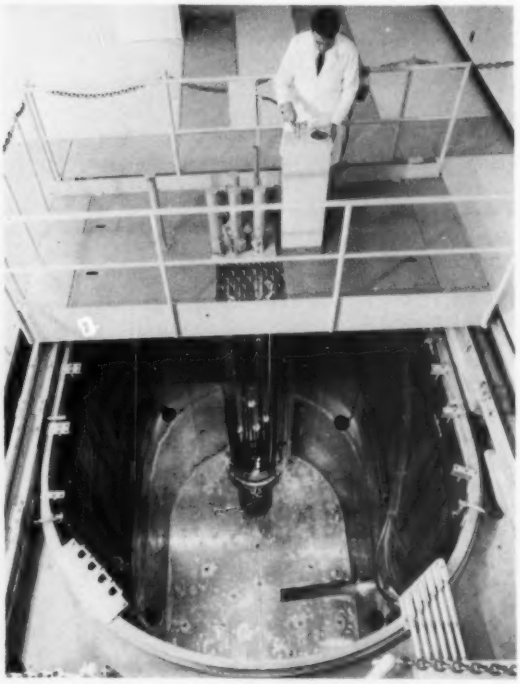

Fig. 5 TRIGA Mark III reactor.

TRIGA reactors use uranium–zirconium hydride fuel elements, which give the core a large prompt negative temperature coefficient. Thus rapid insertion of excess reactivity causes a rapid increase of power, and the concomitant increase in fuel-element temperature both limits the maximum power level and returns the reactor to normal steady-state power levels.

Extremely high transient power levels can be reached with TRIGA reactors that are especially equipped for rapid insertion of excess reactivity. Both the Mark I reactor and the ATPR at General Atomic are equipped for such "pulsing" operation.

A prompt insertion of 2.2% excess reactivity in TRIGA Mark I results in a 1000-Mw pulse with a 15-msec width at half-maximum and provides neutron fluxes of the order of 10^{16} n/(cm^2)(sec). Up to 5 dollars* of reactivity can be rapidly inserted in the ATPR, which gives an 8400-Mw pulse (with 100 fuel elements in the core) with a 5.5-msec width at half-maximum, and provides neutron fluxes of the order of 10^{17} n/(cm^2)(sec). Such pulses can be obtained from 6 to 12 times an hour, depending on the nature of the material being irradiated (high-cross-section samples lessen the repetition rate).

terminates in front of a detector for work with very short-lived isotopes.

As mentioned before, data from the gamma-ray spectrometer at the TRIGA facility can be automatically transferred via a card-punch unit in the activation-analysis building to cards for computer processing.

Linacs. Each of the two General Atomic Linacs (Figs. 6 and 7) is an L-band disk-loaded traveling-wave machine energized by high-power klystron amplifiers.

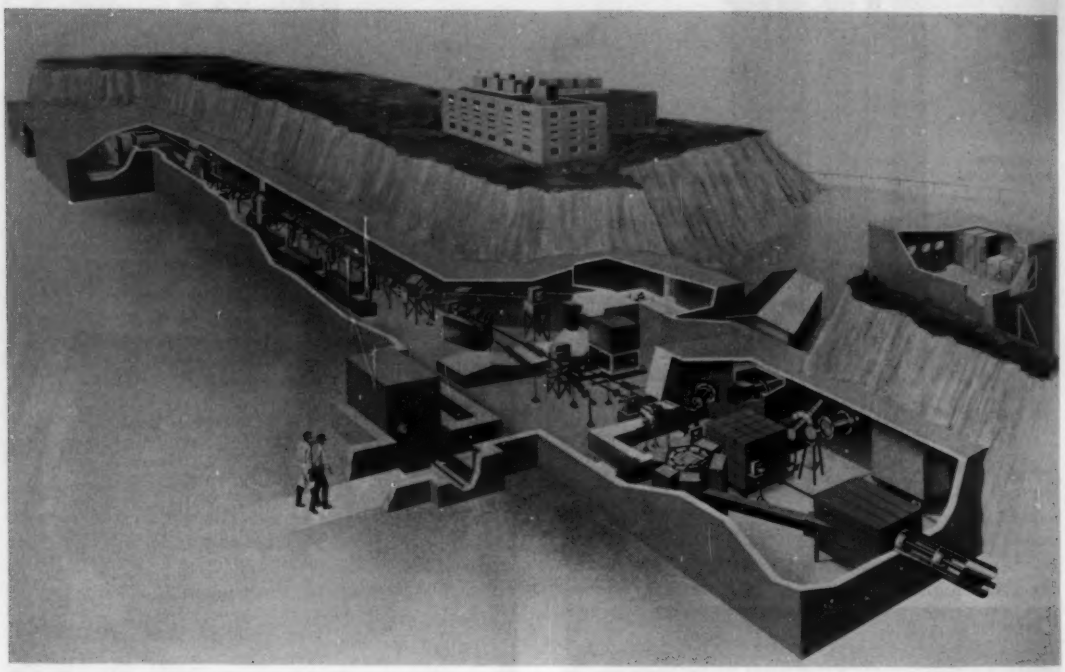

Fig. 6 Linac building, conceptual drawing.

The pneumatic sample-transfer systems at TRIGA run between the reactor outer core position and a special sample-handling room in the TRIGA building. In the room a multichannel gamma-ray spectrometer, which includes both solid and well type sodium iodide detectors, provides for immediate counting of samples after they are removed from the reactor. The system provides for automatic timing of the irradiation period, a delay period before counting, and a counting period. One of the transfer systems

One Linac can be operated to give monoenergetic electron beams at any energy between 1 and 45 Mev in 5-μsec pulses at a repetition rate adjustable from 1 to 180 per second, or 360 per second for pulses lasting 2.5 μsec, or 720 per second for pulses ≤ 0.5 μsec. Pulse durations are adjustable between 0.01 and 5 μsec. Peak currents of 2000 ma are obtainable with the shorter pulses, and 700-ma currents can be achieved with > 0.5-μsec pulses. The average beam power ranges from 4 kw at 8 Mev, up to 13.6 kw at the most efficient energy of 25 Mev, and then back to 4 kw at 40 Mev.

The second Linac provides monoenergetic electrons at any energy between 1 and 17 Mev. Pulse duration

*A "dollar" is the amount of reactivity in any reactor due to the delayed neutrons. Usually it amounts to 0.75–0.85% of the total activity.

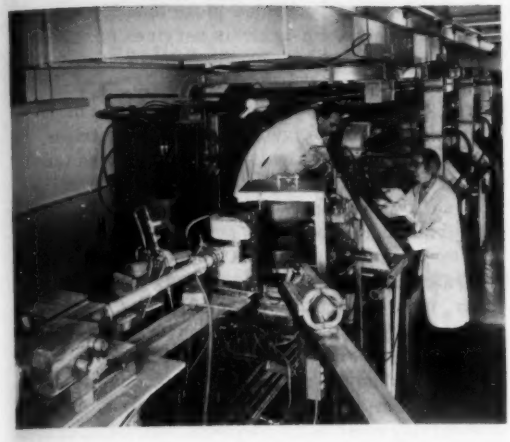

Fig. 7 Linac installation.

and repetition rates are the same as with the larger machine. Average beam power at the most efficient energy of 8.5 Mev is 10 kw.

Fansteel (tungsten alloy) converters may be placed in the path of the electron beam of either accelerator to produce bremsstrahlung for photoactivation-analysis work. Coupling the two Linacs to obtain a 94-Mev electron beam is planned.

A sample transfer tube between the Linac and the activation-analysis buildings is being installed at this time. This will remove the present need for using portable equipment in photonuclear work with short-lived isotopes and will bring the permanent counting facilities of the activation-analysis group into convenient use for photoactivation analysis.

Cockcroft–Walton Accelerators. The C–W bunker consists of two large underground rooms separated by 1-ft-thick concrete walls and 12 ft of earth. Overhead shielding consists of 1-ft-thick concrete and 15 ft of earth. The shielding, which exceeds health physics requirements, provides for mutual isolation of separate neutron experiments at the various sites. Thus the C–W activities do not interfere with neutron time-of-flight experiments at the Linac.

The two Texas Nuclear Corporation C–W's are presently located in one of the two rooms (Fig. 8). They can provide 2.5-ma beams of deuterium ions at 150 kv and are used to generate up to 2.5×10^{11} 14-Mev n/sec.

The two generators supply two completely separate analytical systems; each system includes a generator-control console, sample-transfer tubes, and counting equipment. One system is operated from the activation-analysis building and is used mainly for research purposes. The other is operated from the second underground room and is used for routine analyses.

A completely automatic analytical system is used for routine analyses in the C–W bunker. Samples are placed in a rack from which they are automatically and sequentially put through a preset analytical regimen of irradiation and counting. The system includes the simultaneous irradiation and counting (with a separate counting system) of a flux monitor. Both the sample and flux monitor are spun during the irradiation.

DISCUSSION

The integrated array of equipment described above provides for very sensitive determination of nearly every element. The reactor is the most powerful and generally useful activation tool, but the Linacs and C–W's also have very important roles.

The TRIGA reactors, with their large fluxes of both thermal and fast neutrons, enable the determination of about 75 of the 81 stable elements with an average sensitivity of 1 nanogram; the more sensitive elements can be determined at the parts-per-trillion level in a sample.

The C–W's supply a high flux of fast neutrons only, which provides for the selection of fast-neutron-reaction analyses without interference from thermal-neutron-reaction products.

The Linac makes possible the generation of neutron-deficient products, in general, and the ability to select beam energy makes it possible to avoid certain unwanted reactions.

With these devices, purely instrumental activation analysis (i.e., simply irradiation and counting of the sample) of many complex samples can be carried out since the possibility of promoting analytically useful reactions, while avoiding unwanted reactions, is maximized. For example, iron, nickel, and manga-

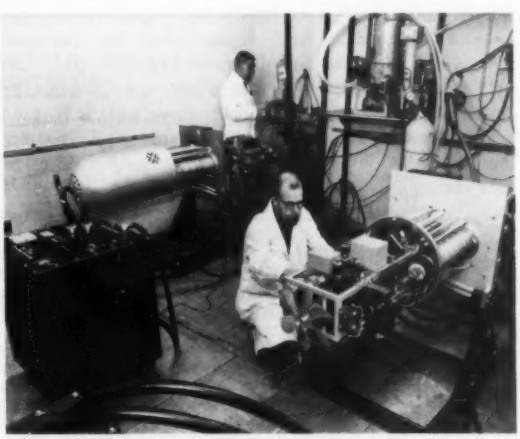

Fig. 8 Cockcroft–Walton installation.

nese are best determined in many soils with the C–W, Linac, and TRIGA, respectively. Iron is determined with adequate sensitivity by the $^{56}Fe(n,p)^{56}Mn$ reaction with the C–W since the other elemental constituents of soil generally have noninterfering combinations of concentration, half-life, and sensitivity. With the reactor or Linac, many elements have much better sensitivity than iron, and interferences are more likely to occur. Similarly, nickel can be determined with good sensitivity by promoting the $^{58}Ni(\gamma,n)^{57}Ni$ reaction (threshold 12 Mev) without undue interference from other elements in soil, whereas neither the reactor nor the C–W would permit an instrumental determination. Finally, the thermal-neutron reaction $^{55}Mn(n,\gamma)^{56}Mn$ provides such a very favorable sensitivity for manganese that its determination in soil with the reactor is straightforward, but neither the Linac nor the C–W can be used conveniently.

Although about 65, 10, and 6 elements are analytically favored by TRIGA, Linac, and C–W irradiations, respectively, in each sample there will be an optimum combination of irradiations for the determination of certain elements. The optimum will include cost and speed factors in addition to analytical feasibility, and the activation-analysis group carries on a considerable research and development effort to determine the best way to approach the myriad of different analyses.

Despite the capabilities of the equipment, it often is necessary to perform radiochemical separations to remove interfering radioisotopes produced during activation of a sample. The majority of the activation-analysis-group personnel are experienced radiochemists, and this phase of activation analysis is under a continuing program of research and development.

General Atomic provides more than 500 organizations with activation-analysis services for elemental determinations. These services have included research and development projects on such subjects as the determination of trace elements in blood, the determination of oxygen in specific matrixes, the use of activation analysis in criminalistics, and the use of reactor fast neutrons in activation analysis. Routine service analyses that are carried out for industrial, university, and government organizations run the gamut of analytical situations, including the determination of trace elements in water, seawater, foods, food extracts, beverages, plants, soils, ores, metals, biological tissues, semiconductors, paints, cements, rubbers, petroleum (crude and refined products), plastics, glass, paper, and other matrixes.

In addition to being a very sensitive means of determining most of the elements, activation analysis is often nondestructive, and in most cases the yield of a given indicator radioisotope is a linear function of the concentration of the parent element in the sample. The latter feature means that an unknown material can be analyzed for its major elements as well as its trace elements, and the General Atomic group has developed an analytical regimen with computerized data reduction to survey samples for their complete composition; firm upper limits are given for elements not observed in the analysis. The survey-analysis program has filled an especially important role in the field of product specification.

The activation-analysis group also offers courses in activation analysis, which many scientists have attended. In addition, a number of scientists from this country and abroad have visited General Atomic for times ranging from a week to a year to use the facilities and obtain advanced training in the field.

(PSB)

MARSEILLE SEMINAR ON USE OF RADIOELEMENTS IN THE CHEMICAL AND PETROLEUM INDUSTRIES

A seminar on the use of radioelements in the chemical and petroleum industries was held June 16 and 17, 1966, in Marseille, France. Sponsored by A.T.E.N. (Technical Association for Nuclear Energy), the meeting covered numerous fields of radioisotope applications. Besides the introductory general session, the program was divided into three parts: nondestructive testing, tracer use, and analytical chemistry applications.

The use of ^{60}Co as a gamma-radiation source for radiography of pipe walls and welds and in thickness, level, density, and moisture gages was discussed.

Several papers were presented on the application of radioactive tracers to finding leaks, studying mixing, measuring flow rates, and investigating corrosion. One paper described generators, or "cows," for preparing short-lived radioisotopes, e.g., 2.6-min ^{137}Ba, for tracer use. Because they do not cause any fire hazard, luminous sources based on tritium were mentioned as of interest for the petroleum industry.

Analytical methods discussed included introduction of stable isotopes into a process and determination later by activation analysis; X-ray fluorescence with isotope sources; and isotope dilution. Various applications of activation analysis were mentioned. Some specific techniques described were: continuous determination of silicon in a $SiHCl_3 + H_2$ mixture by absorption of soft X rays from ^{55}Fe or of bremsstrahlung from tritium absorbed on zirconium or titanium; determination of oxygen by release of a corresponding amount of thallium ions from radioactive metallic thallium; and determination of chlorine by ^{110}Ag-labeled reagent. The program is given in some detail in *Energie Nucleaire*, 8: 333-336 (1966).

(Martha Gerrard)

Section III

Isotopes and Radiation Technology

Food Irradiation

International Symposium on Food Irradiation, Karlsruhe, Federal Republic of Germany, June 6-10, 1966

[*Editor's Note: Isotopes and Radiation Technology* continues its coverage of food-irradiation technology with this report on the International Symposium on Food Irradiation held June 6–10, 1966, at Karlsruhe, Federal Republic of Germany. From the number and variety of papers presented at Karlsruhe, food irradiation seems clearly to be a topic of increasing importance. To some nations it is valuable as a new commercial process; to other nations it may mean survival for their people. Although the United States is a leader in the field and presented the largest number of papers (17), Canada, Japan, and the USSR seem to be pressing ahead with commercialization of the process at a faster pace than the United States. The concern of several authors about ensuring ease of international trade in irradiated foodstuffs was evident; legislation of any kind on food irradiation varies widely. The number of papers simply describing facilities for food irradiation was indicative of the growing importance of food irradiation throughout the world. The papers are reviewed in the order in which they were presented.]

Abstract: The International Symposium on Food Irradiation, Karlsruhe, Federal Republic of Germany, June 6–10, 1966, was sponsored by the International Atomic Energy Agency (IAEA) and the Food and Agriculture Organization (FAO). Representatives from 17 countries presented papers: 6 on radiation sources and dosimetry; 6 on wholesomeness of irradiated food; 8 on the chemical and physical effects of ionizing radiation; 9 on microbiology, virology, and quarantine problems; 17 on the status of various irradiated commodities; 12 on programs and facilities for food irradiation; 5 on economics; and 5 on legislation and clearances of irradiated food.

Goldblith[1] opened the symposium with a historical review of the work on radiation preservation of food. The first successful attempt to use ionizing energy to preserve food was reported by Proctor, Van de Graaff, and Fram.[1a] Much of the early developmental work was done in the United States where the U. S. Atomic Energy Commission (AEC) began supporting research about 1950 and the U. S. Army started in 1953. After Hannan and coworkers in Great Britain and Kuprianoff and coworkers in Germany led early investigators in Europe, a study group on food irradiation was set up in January 1960 by the Organization for Economic Cooperation and Development's European Nuclear Energy Agency — the first of many such international efforts. Canada is now furthest advanced toward commercialization of the process, with the USSR, Japan, and the United States following closely. Goldblith emphasized that wholesomeness studies have always shown that irradiated food is harmless to human beings.

Radiation Sources and Dosimetry

Existing installations in the United Kingdom were described by Jefferson.[2] A 4-Mev linear accelerator has been operating at Wantage Research Laboratory since 1956; the first isotope facility was the Spent Fuel Element Facility at Harwell, which began operating in 1958 at a level of 2 Mc. The Package Irradiation Plant at Wantage, which has been used to treat many foods since 1960, can run unattended for 5 days and nights. The economics of any food-irradiation operation are governed by the cost of radiation, the throughput handled, the required dose, and the radiation utilization efficiency.

Kuhl[3] reviewed ^{60}Co source development at Brookhaven National Laboratory (BNL). Sources were developed as large tubes and flat strips, primarily for research and development. The two standard strips, the BNL Standard Mark I and Mark II, are interchangeable; the Mark II can be reirradiated. The cost of a strip ready to insert into a reactor is about $56. Since measurement of absorbed dose is important to the radiation processing industry, BNL studies and evaluates known dosimetry systems. A summary of data is included in Table 1.

Grünewald[4] presented design data for machine sources. The amount of radiation actually absorbed by the sample, L (Mrads × kg/hr), is equal to $0.36 \times \eta \times P$, where η is beam utilization efficiency and P is beam power (watts). The equation can be modified for isotope sources since 1 thousand curies of ^{60}Co is the equivalent of about 14.8 watts and 1 thousand curies of ^{137}Cs is the equivalent of about 3.6 watts. The efficiency of a machine source depends on the beam utilization efficiency. Soft-X-ray generators have the advantage of variation in dose rate, but higher

Table 1 SUMMARY OF DOSIMETRY SYSTEMS

Dosimeter	Type	Range	Applications and limitations	Ref.	Radiation
Standard Fricke	Integrating	1 to 4×10^4 rads gamma dose	Accepted standard for gamma dose Exposure dose measurement Absorbed dose measurements in any medium	3*a*	Gamma rays and charged particles
Deaerated Fricke	Integrating	Up to 9×10^5 rads gamma dose	Exposure dose measurements Absorbed dose measurements in any medium	3*b*	Gamma rays and charged particles
Ferrous–cupric	Integrating	Up to 9×10^5 rads gamma dose	Exposure dose measurements Absorbed dose measurements in any medium	3*c*	Gamma rays and charged particles
Ultraviolet transmitting (UVT) Lucite	Integrating	10^4 to 3×10^6 gamma dose	Exposure dose measurements Absorbed dose measurements in any medium	3*d*, 3*e*	Gamma rays and charged particles
Blue cellophane dye dosimeter	Integrating	1 to 6×10^6 rads gamma dose	Exposure dose measurements Absorbed dose measurements in any medium	3*f*	Gamma rays and charged particles
Thimble ionization chamber	Dose rate	Up to 10^6 r/hr gamma rays	Exposure dose measurements Absorbed dose measurements in any medium	3*g*	Gamma rays
Victoreen condenser ionization chambers	Integrating	Up to 250 r gamma rays	Calibration standards Exposure dose measurements Absorbed dose measurements in any medium	3*h*	Gamma rays
Victoreen roentgen rate meter	Dose rate	Up to 5×10^4 r/min gamma dose	Exposure dose measurements	3*h*	Gamma rays
Anthracene scintillator	Dose rate	0.1 to 1 r/hr gamma dose	Exposure dose measurements Absorbed dose measurements in any medium		Gamma rays
n on *p* solar cell	Dose rate	Up to 10^7 r/hr gamma dose	Exposure dose measurements Absorbed dose measurements in material having density of 2.0 g/cm^3 only Area monitoring Source comparisons	3*i*	Gamma rays
p on *n* solar cell	Integrating	10^4 to 10^8 r/hr gamma dose	Exposure dose measurements Area monitoring, can be used above 200 kev only	3*j*	Gamma rays

power X-ray generators give deeper penetration. For both types, dose distribution depends on the distance between the sample and the window of the unit, with the evenness indirectly dependent on conveyor-belt velocity, beam diameter, and scanning frequency. The difference between mean dose rate and instantaneous dose rate, which can be much higher, is especially important for biological systems.

The development of equipment with a very high X-ray output was described by Rajewsky.[5] Originally developed for investigating problems in radiation biophysics and biology, this equipment can easily be used to irradiate food at dose rates up to 10^7 r/min.

Dvornik, Zec, and Ranogajec[6] developed a new dosimeter for high levels of radiation based on their discovery that, with ^{60}Co, the G value for radiolytic hydrogen chloride from chlorobenzene solutions in ethanol was 5 ± 0.1 and was independent of dose (0.05 to 100.0 Mrads), dose rate (0.5 to 2500 rads/sec), and chlorobenzene concentration (4 to 40%). The hydrogen chloride concentration in the whole dose interval is determined to within 1% by mercurimetric or alkalimetric titration directly in the irradiated sample. Similar methods are of possible interest for calibration and control of industrial irradiation with intense electron beams.

The first session ended with a paper by Chadwick[7] on the distribution of radiation energy in rotating fruit. His results indicate that rotating fruit as it is irradiated shifts the peak of the energy distribution nearer the surface of the fruit. He discussed the effect that the change in energy distribution might have on the choice of dose and called for a clear and standard definition of electron dose in food irradiation. Food technologists must decide whether the dose is to be measured at the surface of the fruit or at the layer receiving peak intensity.

Wholesomeness of Irradiated Food

Hickman[8] presented results of feeding tests carried out in the United Kingdom. No adverse effects attributable to consumption of irradiated food appeared in either short-term studies with rats, chickens, and pigs or in long-term studies with rats and mice. The animals ate grain, eggs, ham, fish, horsemeat, and potatoes treated with pasteurizing and sterilizing doses of radiation. Microbiological assays showed that vitamin losses were of the same magnitude as those caused by thermal methods of food preservation. No effects on the nutritional value of protein were found. Raica and Howie[9] reviewed the U. S. Army program on wholesomeness of irradiated food. Data have been obtained through the efforts of over 30 academic, commercial, and government laboratories; over 20 different foods were fed to 15 thou-

sand mice, 5 thousand rats, 250 dogs, and 37 monkeys. Irradiated foods have been demonstrated to be as wholesome and, in general, as acceptable as conventionally processed foods. Whitehair[10] reported that none of the studies by the U. S. Army Surgeon General's Office on 21 irradiated foods had shown any of them to be unwholesome. Of great current interest are reports that suggest that degradation or breakdown products might have mutagenic or cytotoxic effects in man; well-designed in vivo experiments are needed to investigate this problem.

Lang and Bässler,[11] from the University of Mainz, gave the results of feeding irradiated fats to rats. Oils irradiated with 2.5 Mrads caused no detrimental effects after 80 weeks of feeding, although oils irradiated at higher levels had various ill effects. Animals fed oil irradiated at 50 Mrads lost weight after 24 weeks and died in less than 75 weeks, about half a normal life span. Oil irradiated at 100 Mrads produced immediate toxic symptoms in the animals. The dimerization of fatty acids is assumed to be the cause of damage since there is no indication of peroxide formation. Intensive studies with pigs were undertaken by Dammers et al.[12] on the effects of feeding untreated meal, heat-treated meal, and irradiated meal (1 Mrad from ^{60}Co). The results indicated that neither heat nor radiation produced undesirable nutritional effects in feed for pigs. The absence of *Salmonella* in the feces of pigs receiving treated feed demonstrated the efficiency of the treatments and the efficacy of the method to prevent the occurrence of healthy *Salmonella* carriers among pigs.

Because only 7 to 15 krads are necessary to inhibit potato sprouting, many of the problems associated with other food-irradiation studies are missing from potato feeding studies. Lang and Bässler[13] reported feeding rats on diets of a mixture of 72% dried potatoes, 15% casein, 10% Mazola oil, and 3% salt. One group eats unirradiated potatoes; another group eats potatoes irradiated at 10 krads. No differences have been noted so far.

Chemical and Physical Effects of Ionizing Radiation

The wheat component most severely affected by radiation is the polysaccharides, reported Deschreider.[14] Gamma radiation generally reduces proteolysis in flour, and high doses cause the wheat proteins to become partially denatured as well as polymerized and condensed. Radiation doses below 150 krads can improve baking properties because they maximize starch viscosity, gliadin solubility, the Hagberg index, and substances that can be precipitated by electrodialysis.

Thieulin, Basille, and Morre[15] have been trying to develop means of identifying irradiated food on the basis of recognizing chemical effects. They investigated 16 simple sugars, monosaccharides, and their derivatives, paying special attention to galactose. At *p*H 9.5 and 0.001*M* concentration, borated sugars are 2 to 12 times as sensitive to radiation as pure sugars are; maximum sensitivity is obtained with equimolar mixtures (0.1*M*).

Merritt[16] reported using programmed cryogenic-temperature gas chromatography and rapid-scanning mass spectrometry to successfully identify the compounds that give irradiated meat its characteristic odor and flavor. The odor is the same for beef, pork, lamb, and other meats; it varies only in intensity. The odor, which is a direct result of the radiation, is reproducible for a given radiation dose and is not affected by water, oxygen, the absence of oxygen, or any such variable in the radiation environment. Since carbonyls, alcohols, thiols, thioalkanes, and esters are also found in unirradiated meats, they are not the guilty compounds. New analytical techniques have shown that homologous series of *n*-alkanes and *n*-alk-1-enes are found in irradiated meat but not in appreciable amounts in unirradiated meats. The mechanism of irradiation damage in lipids now seems clearly to be radiation-induced direct bond cleavage, which forms primarily alkyl free radicals. In glycerol stearate, for example, scission of the bonds at all points of the chain with recombination or hydrogen termination of the resulting alkyl free radicals would form the *n*-alkanes from methane to heptadecane. All alkanes from methane to hexadecane are found in good yield.

The origin of the various components in meat volatiles was determined by analyzing meat protein and meat fat separately. A 500-g sample was separated into a protein, a lipid, and a lipoprotein fraction; the fractions were irradiated separately and analyzed. Since the lipoprotein fraction produced aliphatic hydrocarbons and sulfur compounds and it was the only fraction with the characteristic irradiation odor, the alkanes were clearly at fault. The data thus far support the simple hypothesis that radiation products are the result of direct bond cleavage. The main products of irradiation of dry oxygen-free lipid substances are the *n*-alkanes, *n*-alkenes, and traces of *n*-alkynes. Sterols give primarily normal alkanes and isoalkanes from cleavage of the alkyl side chain. Proteins and peptides show little evidence of rupture of the peptide bond, and the main products are from cleavage of the side chains on end groups. The amino acids with aromatic rings or with sulfur groups tend to be most radiation sensitive.

A novel method of estimating the state of preservation of fish was proposed by Turjanski and Kramar,[17] who used electrode-carrying pincers and a tester commonly used in electronics to measure both capacitance and resistance of fish. They decided that capacitance is the variable more closely associated with the state of preservation.

Dubois et al.[18] reported the results of analyzing the volatile substances in irradiated apple juice. Acetaldehyde, isobutyraldehyde, isovaleraldehyde, butyraldehyde, and caproaldehyde were produced in normal apple juice by irradiation, but only the first three were produced in concentrated apple juice. Grape juice irradiated at 0.1 Mrad gave off a volatile disulfide that is probably responsible for the characteristic odor of irradiated juice.

Irradiated wheat and flour prepared from it were analyzed by Pahissa-Campa et al.[19] Doses from 0.01 to 2.0 Mrads were used. Differences were noted in the texture, odor, color, and taste of bread made from wheat that had received the highest dose.

Tuchscheerer[20] described experimental methods of analysis to determine the chemical composition of food for use in making predictions about induced radioactivity. From 0.05 to 10 Mev, the biggest problem is stimulation of isomers by (γ,γ') reactions, with ^{107m}Ag being most troublesome. Above 10 Mev, (γ,n) reactions are the problem. A new simplified method of calculating induced radioactivity was introduced, and the results were compared to experimental values. The equation uses the summation of a set of integrals. When average European food was irradiated and maximum radioactivity determined, a waiting period of only 5 days was enough to reduce the induced radioactivity below the permissible level for ingestion.

Radiation doses sufficient for microbiological stabilization are frequently insufficient to prevent enzymatic deterioration. Vas[21] reviewed the known data on this problem. Present evidence indicates that the decrease in enzyme activity is a negative exponential function of the dose. Vas feels that most of the contradictory quantitative data on inactivation dose values could be accounted for if the purity of the enzyme were known. It is still unclear whether irradiation inactivates dissolved enzymes by causing aggregation and thereby removing the enzyme from its normal sphere of activity or by directly damaging the active enzyme surface. Enzymes are more sensitive to radiation when an animal is under stress; so stress is being generated in animals by a low whole-body dose of radiation or by an adrenalin injection 24 hr before slaughter.* Methods combining radiation, heating, refrigeration, and physiological methods might be used.

Microbiology, Virology, and Quarantine Problems

Ingram[22] reviewed the three terms proposed by an international committee.[22a] *Radappertization* will mean radiation sterilization to prevent microbial spoilage; *radicidation* will mean radiation pasteurization to control organisms of significance to public health; and *radurization* will mean the use of radiation to prolong shelf life. Ingram also reviewed the important microbiological objectives of irradiation. Radappertization's goal is to control *Clostridium botulinum*; however, to reduce the population by a factor of 10^{12} generally requires 5 Mrads and causes unfortunate organoleptic changes. Attempts to modify the effect of the dose have entailed using radiation sensitizers, adding activated charcoal, and irradiating food in the frozen state. The two basic difficulties attending all radicidation proposals have been deciding how much radiation is necessary and how the treatment is to be controlled in commercial application. Radurization depends on the more or less complete destruction of spoilage microflora, but, to minimize risk to public health, a somewhat smaller extension of storage life should be accepted so that the spoilage microflora left could indicate when the food is spoiled.

The resistance of *C. botulinum* type E to radiation was covered by Segner and Schmidt,[23] who conducted studies with a 0.1-Mrad dose at 35°F in haddock homogenate and in neutral phosphate buffer. Semilog survivor curves for spores of the Beluga, 8E, Alaska, and Minneapolis strains were characterized by a pronounced lag in reduction during the first cycle followed by exponential destruction. The dose for 90% reduction from the exponential portion of a survivor curve (D value) was 0.22 Mrad in the haddock and 0.08 to 0.11 Mrad in the buffer at 46°F. Stehlik and Kaindl[24] combined heat and radiation to reduce *Saccharomyces cerevisiae* var. ellipsoideus. The combination was more effective than heating before or after irradiation. A synergistic effect of about 2 orders of magnitude showed up for the combination of temperatures above 104°F and doses of about 0.15 Mrad.

Grecz[25] reported that D values varied with temperature, medium, and size of spore inoculum. Temperature affects radical formation, chemical reactivity, extent of annealment, and radical diffusion. Interaction of these effects can explain why spores are most sensitive at 32°F. Temperature also affects the DNA directly, although the effect depends on the vibrational state and extent of hydration of the DNA molecules.

Turjanski et al.[26] irradiated dried cattle albumin at doses from 0.25 to 4 Mrads and reported sterilization of red cells at 0.5 Mrad and white cells at 2 Mrads. Efforts will continue toward defining the biochemical behavior of the albumin.

Massa[27] presented the results of attempts to inactivate foot-and-mouth disease virus by irradiation. A dose of 3 Mrads in the dry state and 4 Mrads in the liquid state will inactivate cultures in vitro, re-

*A recent report in *New Scientist* (7 July 1966), page 18, suggests that the opposite effect is necessary in fresh pork. Some researchers have proposed injecting a tranquilizer before slaughter. — *The Editor*

ducing the number of viruses by a factor of 10^7. To inactivate the virus in situ requires 2 Mrads for blood and bone marrow and 1.5 Mrads for lymphatic glands. Sterilization of carcasses is feasible, but high doses are necessary and will probably have undesirable organoleptic effects.

Data accumulated over the last decade indicate that salmonellosis is becoming a major health problem. Ley[28] discussed the use of radiation to control *Salmonella* in frozen egg, frozen meat, and dry animal feeds. An inactivation factor between 10^5 and 10^7 is necessary, and a dose between 0.5 and 1.0 Mrad is required. Since the irradiation of liquid whole egg produced undesirable organoleptic changes, an effective and cheaper heat-treatment was developed for this food. A dose of 0.45 Mrad effectively treated desiccated coconut, but a small flavor change and a slight darkening were deemed unacceptable by the trade. A dose of 0.4 Mrad for broilers and ducks frozen at −4°F did not affect the quality of the birds and was microbiologically effective. Imported horsemeat from Argentina and kangaroo meat from Australia for pet food can be treated by a dose of 0.65 Mrad at a cost of about 0.6 cent/lb. The entire state of the art of using ionizing radiation to radicidate *Salmonella* was covered by Mossel.[29] Staple protein foods of low water activity[29a] were chosen for the first applications of radiation to food processing because of (1) restricted radiochemical damage to the commodities, (2) absence of microbial proliferation after irradiation, and (3) technological advantages such as packaging. After laboratory-scale tests showed promise, pilot-plant-scale tests confirmed that a dose of 0.7 ± 0.1 Mrad eliminated *Salmonella* from frozen and dried chickens, fish meal, and mixed feed. Studies with rats and piglets have shown that there are no effects from eating irradiated foods. The third step, tentative commercial-scale experiments, is proposed for countries that export large quantities of foods susceptible to *Salmonella*.

Cornwell[30] discussed the import–export uses of radiation. The great increase in international travel and trade, and the speed of movement from one part of the world to another necessitates strict measures to prevent worldwide dissemination of unwanted insects. Irradiation can help in two ways: irradiation at port of entry, and use of the sterile-male technique. Equipment is in the planning stage for irradiation of passenger baggage going to the mainland United States from Hawaii; a pilot-scale evaluation will be performed in 1967. The damage that repeated irradiation may have on items often carried in passenger baggage will be studied; tourists will, of course, have to remove their cameras and film. The distribution of fruit-fly species in Australia prevents marketing of fruit between states; New Zealand requires that fruit be stored 2 weeks at 31°F before it can be imported. A plant to irradiate timber and lumber to disinfest wood wasps and the European house borer would cost about 0.6 cent/cu ft; present fumigation treatments cost about 1 cent/cu ft. To prevent spread of the Mexican fruit fly, sustained release of sterile males along the Mexican border is thought to be more economic than the current practice of quarantine survey and insecticide spraying, which costs about a hundred thousand dollars per year. On the other hand, control of the Queensland fruit fly in Australia is not considered practical because of the large area of infestation.

Status of Various Irradiated Commodities

This was the largest session of the conference, with twice as many papers as in most other categories. Urbain[31] pointed out that, economically, radiation is not yet competitive with refrigeration for meat and poultry. Consumer acceptance has not been determined, and facilities for producing sufficient radiation-sterilized meats to test consumer acceptance are not readily available. Hansen[32] emphasized that radiation will not be used commercially unless it results in a better product or is cheaper. Radiation-produced off-flavors are a serious hindrance to acceptance of irradiated meat. Danish work has concentrated on dose distribution and dose rates in canned meat.

Work on poultry and eggs was covered by Thornley.[33] Radicidation at doses of about 0.5 Mrad will control *Salmonella* in frozen whole egg. Radappertization of poultry carcasses is achieved by doses of 4.5 Mrads, but other processes must be used with irradiation to prevent undesirable off-flavors. The most valuable commercial process is radurization of poultry; doses of 0.15 to 0.25 Mrad extend storage life at temperatures below 41°F by a factor of 3.

Saint-Lebe, Guilbot, and Pelegrin[34] combined gamma radiation and slow drying by cool ventilation to replace the standard technique of treating damp harvested maize by rapid drying with hot air. They reported that radiation doses of less than 0.5 Mrad had no effect on the extraction yield of starch from maize, whereas drying at 165°F lowered the yield about 4%. The combined treatment was done on a laboratory scale under experimental conditions easy to transfer to the industrial level. The radiation effect on mycoflora persisted 26 days, and the first mycoflora regenerated was most sensitive to the dry air flow reaching it.

Control of insects in grain by irradiation was discussed by Cornwell.[35] The effective dose for sterilizing the grain weevil *(Sitophilus granarius)* is 16 krads; late pupae are still able to produce adults, but these die soon after emergence. Substerilizing doses reduce irradiation costs appreciably and control but do not wipe out infestation. The 16-krad dose will sterilize or kill nearly all known cereal-infesting

insects, including the khapra beetle. High temperatures (86°F) before irradiation sensitize grain weevils to killing by radiation, but high temperatures during irradiation make them resistant, and high temperatures after irradiation merely increase the mortality rate. Temperature does not affect the weevil's resistance to sexual sterilization. Grain carbohydrates are unaffected nutritionally even at high doses, and the biological value of maize protein and wheat gluten is unaffected at doses up to 2.8 Mrads. Radiation has little effect on the B-complex vitamins, although a 10% loss of thiamine has been reported at 50 krads. Assays in the United Kingdom have shown little destruction of the supposedly radiation-sensitive vitamin E at doses of 0.2 Mrad. Doses many times higher than 16 krads may be used without causing changes of commercial significance, always excepting emotional reactions of consumers; organoleptic appraisals have shown no effect with the proposed dose. Practical and economical considerations suggest that gamma irradiators can handle up to 100 tons of grain/hr, and accelerators can handle up to 400 tons/hr. Neither method is likely to compete with fumigation at annual throughputs of less than 100 thousand tons. The technique is most likely to be exploited in those countries that consume most of their grain locally; it is least likely to be used in those countries that export grain.

Golumbic and Davis[36] reviewed the use of radiation to treat insects. Over 50 years ago Morgan and Runner[36a] tried to control the cigarette beetle, *Lasioderma serricorne*, in packaged cigars with X-radiation; they failed, but since then a wealth of data has been accumulated. The current objective of controlling insect infestation with radiation is the prevention of storage losses and extension of storage life of grain. The most complete summary of the action of ionizing radiations on insects is by Hilchey.[36b] Two generalities are that lethal doses for insects are high compared to lethal doses for warm-blooded animals and that there are great differences between species as well as between development stages. Very high doses are needed to sterilize moths as compared to beetles in stored products; in most species the female is more susceptible to radiation than the male. Four questions that need to be answered for irradiation of packaged products are: (1) What dosage level will give 100% mortality of stored-product insects? (2) What is the attenuation coefficient of the package and product? (3) What are the effects of radiation on food and packaging materials? (4) How feasible is the use of radiation in packaging lines for processing food? The studies that report the use of radiation to kill fungi associated with seeds indicate that radiation is not practical because of its effect on germination.

The papers on radiation treatment of fish and seafood began with Shewan,[37] who pointed out that radiation sterilization has consistently produced undesirable organoleptic changes in marine products, even though about 20 species of fish and shellfish respond favorably to radiation pasteurization. Shelf life can be extended 1.5 times at 40°F and 2 times at 32°F by such treatment. Maximum extension of shelf life depends on the length of time between catching the fish and irradiating it; ideally, a shipboard irradiator should be used. Present studies indicate that the problem of *C. botulinum* type E is under control.

The status of research and development of radiation pasteurization of fish in the United States was reviewed by Slavin, Ronsivalli, and Connors.[38] Doses of 0.15 to 0.45 Mrad can double the refrigerated shelf life of the major species of fish marketed fresh or frozen in the United States. Major variables are storage temperature, quality of raw material, and dose level. The use of either bactericidal salts or heat with radiation achieves a synergistic effect. Sinnhuber et al.[39] sterilized sausages made from comminuted fish, corn meal, and gelatin. Patties made from the sausages were canned and then irradiated at 4.5 Mrads. Taste panels found the patties very acceptable after 12 months' storage at 72°F.

Because the Federal Republic of Germany had restricted the use of hexamethylenetetramine as preservative, Schönborn, Kinkel, and Hafferl[40] investigated the use of radiation to preserve marinated herring fillets. This product is usually matured under the action of acetic acid and salt and marketed in a spiced brine containing about 3% salt and acetic acid (*p*H 4.0). The taste panel felt obliged to try a large quantity of the product before giving its opinion. Although a dose of 0.18 Mrad extended shelf life by a factor of 3 at 59°F, the product was organoleptically unacceptable. Experiments resulted in a combined treatment using 3.3 g of ascorbic acid and 3.0 g of sorbitol per liter of fish plus pickle, addition of dry spices, and irradiation at −22°F. This extended shelf life by a factor of 3. No off-flavors were noted at 0.14 Mrad and only slight off-flavors at 0.21 Mrad.

Kardashev[41] presented results of radiation sterilization of vacuum-packed fish products. During storage the fat in irradiated fish is damaged by oxidation, and colloidal senescence of albuminous tissues accompanied by syneresis occurs. The researchers tried lowering the *p*H and introducing carotenoids and ascorbic acid by adding vegetable dressings and sauces containing tomato paste and vinegar. Pieces of fish cooked until ready to eat, vacuum sealed in glass jars, and irradiated at 1.5 Mrads were sterilized. No changes in the quality of the product were detectable at the end of a year.

Sommer and Maxie[42] reviewed recent research on irradiation of fruits and vegetables, emphasizing the use of radiation as a fungicidal treatment and the physiological response of commodities to radiation.

Radiation is feasible for disease control if (1) the disease causes large losses; (2) alternative methods are ineffectual; (3) commodities are diseased at harvest; (4) properly irradiated foods are safe and free from objectionable odors and flavors; (5) texture changes do not adversely affect a fragile commodity unable to resist the rigors of transit; (6) commodities are not destined for extended storage; and (7) commodities can be treated without excessive handling. However, the lowest dose for effective fungicidal use is noticeably harmful to nearly all commodities. The most promising treatment is the combination of heat and radiation which has been shown to have a 5- to 10-fold synergistic effect. Irradiation of strawberries at 0.2 Mrad is a promising application, but stone fruits (peaches, cherries, etc.) are softened by the same dose. Citrus fruits do not respond well, and pome fruits (apples and pears) are adequately treated by chemicals.

French research was reported by Vidal.[43] Irradiation seems capable of halting germination of tubers, bulbs, and rhizomes; preserving fruits and vegetables in conjunction with freezing; and disinfesting certain commodities. Radioisotope sources would be mainly used, and economic evaluations seem to indicate that the process is competitive. However, Staden[44] reported mostly negative results from the Netherlands. Gamma rays cause a complete breakdown of the tomato's internal structure; neck rot in the onion is not inhibited; and sauerkraut is discolored. Results with irradiated mushrooms were moderately good, but kale irradiated at doses higher than 1 Mrad is organoleptically unacceptable.

Farkas, Kiss, and Andrássy[45] reported a dose of 0.8 Mrad was necessary to destroy or substantially inhibit mold on red peppers; however, that dose had deleterious effects on the peppers. Very low doses did shorten the after-ripening period and increase the pigment content. Dennison and Ahmed[46] reviewed efforts to reduce the $13.8 million annual spoilage loss to the citrus-fruit industry. Doses of 0.25 Mrad in combination with low-temperature storage can extend shelf life of oranges nearly 4 months. No objectionable off-flavors were produced in lyophilized pure orange juice in pellet form irradiated at 0.32 Mrad. Doses sufficient to prevent spoilage in lemons caused skin discoloration, and lower doses accelerated ripening.

Irradiation of tropical fruits and vegetables was discussed by Dharkar and Sreenivasan.[47] The shelf life of mangoes is extended 6 days by a dose of 25 krads in air, nitrogen, or carbon dioxide. Radiation apparently inhibited chlorophyll disappearance in fruit skin and carotenoid formation rather than inhibiting ripening. When the fruit was coated with an emulsion of acetylated monoglyceride, physiological damage resulted from inhibition of respiration. The same low dose did inhibit ripening for about 5 days in guavas, sapotas, and tomatoes but not in bananas, chilies, and oranges. Irradiation of 0.4 Mrad and heating at 158°F for 10 min resulted in good-quality canned mangoes, guavas, sapotas, and apples. Canned peas can be sterilized with 0.8 Mrad and 212°F for 5 min; orange juice was sterilized by 0.4 Mrad and 122°F for 5 min.

Programs and Facilities for Food Irradiation

Fowler, Shea, and Dietz[48] in reviewing the AEC food-irradiation program said that the Marine Products Development Irradiator, the Mobile Gamma Irradiator, and the Grain Products Irradiator are either completed or nearing completion. The Hawaii Development Irradiator, a semiproduction facility, was also described. The U. S. Army food-irradiation program was discussed by Mehrlich.[49] Lévêque[50] pointed out that there are no facilities in France engaged solely in radiation treatment of foodstuffs. The Centre d'Études Nucléaires de Saclay has a 6-thousand-curie source, which is used part time. The Centre d'Études Nucléaires de Cadarache has a 2-thousand- and a 12-thousand-curie source, the latter used as a grain irradiator. The Centre Lyonnais d'Applications Atomiques has an industrial plant whose swimming-pool facility now holds an 80-thousand-curie source, but has a 2-Mc capacity. France also has a Saclay-based mobile irradiator that has a 175-thousand-curie ^{137}Cs source. All these are available for any radiation treatment, not just for food.

The international project at Seibersdorf, Austria, to irradiate fruits and fruit juices was covered by Kaindl.[51] Fruit juice is a model substance for fundamental studies because it has economic value, its components are easily separated and are representative of many foods, and its microorganism of interest is yeast, which has already been studied extensively. The fermentation of yeast in grape juice is not reduced immediately but only after several generations, i.e., 1.8 Mrads stops the colony-forming process after 20 days. A combination of heat (122°F) and radiation (0.15 Mrad) reduced survival by 5×10^4 to 10^5. Results show that apple juice from low-quality mixed varieties can be stored without fermentation for more than a year at room temperature after being heated to 122°F and irradiated at 0.3 Mrad; for grape juice a dose of 1 Mrad is needed.

The extensive work done to commercialize irradiated food in Canada was reviewed by Warland and MacQueen.[52] Atomic Energy of Canada Ltd. (AECL) began potato sprout inhibition studies in 1956, and the first food and drug clearance from the government was obtained in 1960. A mobile irradiator mounted on a fully equipped road trailer spent the 1961–1962 season irradiating 1 million pounds of potatoes near production sites in eastern Canada;

the first commercial food irradiator was designed and built by AECL for Newfield Products, Ltd., in 1965. Future prospects are encouraging for other installations.*

De Zeeuw and van Kooy[53] of Euratom felt that the main obstacle to commercialization of food irradiation is not consumer indifference but industry awareness of the effect on agriculture, fisheries, food-processing industries, packaging and packaging materials, storage facilities, transportation, and other industrial aspects. Another difficulty is that responsibility for applying the technology currently rests with the various atomic energy organizations rather than with the food technological institutes, who should take over the responsibility at once. To help overcome these and other difficulties, a pilot plant to be built at Wageningen has been designed. An 85-thousand-curie ^{60}Co source and a 3-Mev Van de Graaff accelerator will provide the radiation, although the plant is designed for a 250-thousand-curie source. An initial throughput of 500 kg/hr at 0.25 Mrad is expected; the irradiation temperature can be adjusted from ambient to −4°K. Construction was scheduled for July 1966, and operation should commence about 16 months later.

The ^{60}Co facility at the Reactor Centre Seibersdorf (Austria) was described by Weidinger and Kaindl.[54] There are two irradiation chambers, containing 30 thousand and 10 thousand curies, respectively; the first is used to irradiate fruit juice, and the other is multipurpose. The 30-thousand-curie source has a 3- by 3- by 3-m irradiation chamber, a binocular observation system, and a temperature-control system. The smaller source has a 3- by 3- by 3.5-m irradiation chamber and a monocular observation system.

Israel's program is coordinated by a committee consisting of representatives of the Israeli AEC, the Ministry of Agriculture, and the various growers' and marketing associations, Kahan, Eisenberg, and Lapidot[55] reported. The main irradiation facility has a 30-thousand-curie source that can handle up to 50 kg at up to 0.8 Mrad/hr. The effects of growing conditions, maturity, irradiation conditions, and storage temperatures are being investigated for citrus fruits. An insect-control program releases 7 million irradiated Mediterranean fruit flies each week. Potato and onion sprouting inhibition is being studied, and sugar beets are being irradiated to prevent loss of sucrose between harvesting and extraction. Dehydrated vegetables are being irradiated to reduce cooking time; filled chocolate confectionery is being irradiated to control insect infestation; and canned orange juice is being sterilized by radiation.

Matsuyama[56] announced the founding of the Japanese Research Association for Food Irradiation; this organization is expected to play an important role in promoting the national program on development studies and their practical applications. About 30 sources in the kilocurie range and an equal number of electron generators are available in Japan for irradiation studies, and research programs have been extensive. Of particular interest is the fact that quality changes in rice after irradiation with doses of up to 20 krads become statistically insignificant after 8 months' storage. The chemical effects of radiation have been studied in marine algae to improve the quality and yield of extracted agar; aging of alcoholic beverages has also been attempted.

Israel's IRR-1 enriched-fuel swimming-pool type reactor contains a gamma cell for irradiation studies. Lapidot et al.[57] described the 30-thousand-curie source that runs on a carriage at the bottom of the reactor pool. The source plaque completely covers the aluminum window between the pool and the irradiation cell; this geometry allows a "source-to-target" overlap. Dose homogeneity within ±20% is attained inside commercial cases 43 by 31 by 31 cm or sacks 80 by 45 by 45 cm and within ±10% in flat boxes 40 by 20 by 5 cm. Any desired dose rate less than 100 thousand rads/hr can be provided.

India's food-irradiation program is extensive, according to Kumta and Sreenivasan.[58] Radiation has been used to delay ripening in tropical fruits, sterilize fruits and vegetables in combination with heat, extend shelf life of semidried fruits, and inhibit sprouting in potatoes and onions. Radiation of Bombay duck, a locally popular fish that is not amenable to conventional processing, augmented storage life to about 45 days at 35 to 37°F. Semidried Bombay duck and shrimp lasted 90 days at room temperature after a dose of 0.5 Mrad. Lethal doses for common insect pests have been determined. India's research facilities are limited to several ^{60}Co gamma cells, but efforts are under way to set up a food-irradiation and -processing laboratory. The facility would include a 100-kilocurie package irradiator and a 27.8-kilocurie portable irradiator.

Rogachev[59] reviewed the use of radiation to treat food in the USSR. The government has approved potato sprouting inhibition, and work on irradiation facilities started in 1965. Dried fruits and vegetables, food concentrates, and wheat have been disinfested with radiation. Optimum doses have been determined for fresh fruit, berries, and vegetables to lengthen shelf life and inhibit microflora without affecting the commodity. During 1964 and 1965, large batches of strawberries, raspberries, apricots, peaches, grapes, pears, and tomatoes were irradiated. Railway dining cars have been serving experimental consignments of irradiated meat since 1965, and large amounts of

*For more recent information on this facility, see the "Miscellaneous" section of this issue, page 203.

irradiated meat were expected to be served and sold in restaurants and stores in 1966.

Economics of Food Irradiation

Baines and Mosely[60] provided some actual costs of plant construction (Table 2) for a grain irradiator using ^{60}Co. One of the more obvious conclusions from the radioisotope-vs.-machines "war" is that radioisotopes are better for thick packages and electron accelerators for thin films. Comparison of the two types of sources for the grain irradiator under discussion indicated that 360 thousand curies of ^{60}Co will be required for a 16-krad dose at a throughput rate of 50 tons/hr. At this rate, cost is almost independent of source, although cobalt is slightly cheaper. At a rate of 100 tons/hr, ^{60}Co irradiation is more expensive at low load factors but is the same order of magnitude at high ones. In the comparison each plant is designed for maximum throughput, and both sources were amortized over 10 years at 6% to account for plant obsolescence even during downtime. Maintenance time was 300 hr/year for the 3.0-Mev accelerator.

Killam et al.[61] pointed out that generalizing about the prospects for food irradiation is useless. Successful and profitable marketing of irradiated foods will depend on customer acceptance and on favorable cost-vs.-benefit relations. Lévêque[62] stressed that the basic hypotheses of cost evaluation need to be reexamined since efficiencies and load factors can be observed in plants now operating. The continually dropping price of radioisotopes was emphasized.

Lapidot et al.[63] gave the results of a technological economic survey of radiation-preserved agricultural produce in Israel as of July 1965. Irradiation costs were estimated on the basis of available and extrapolated data for small irradiators in the growing areas and large irradiators in the seaports or along main highways. A detailed survey of potato and onion sprouting inhibition showed that current expenditure could be reduced from 15% to about 8% of the total crop value with a net return of about 24% on the necessary investment.

Rindorf[64] carried out a study for an industrial concern that decided to establish a multipurpose irradiation plant in Denmark using a Linac. Prices are lowest only when a plant can run continuously for a high percentage of hours a year; some industrialists doubt that they could reach a satisfactory number of hours. The percentage breakdown of costs arrived at is as follows: Linac and attachments, 45 to 50%; land and site work, 5 to 10%; buildings and shielding, 10 to 15%; equipment, 10 to 15%; and sundry expenses, 20 to 25%. For a 50-kw accelerator of 5 to 10 Mev used to irradiate goods between 0.5 and 4.0 cm thick for about 6000 hr/year at a dose of 1 Mrad, 50% utilization, and amortization over 5 years, the cost is between $750 thousand and $1 million.

Table 2 COMPARISON OF COSTS FOR A GRAIN-IRRADIATOR PLANT

Radiation source	3-Mev accelerator			^{60}Co gamma irradiator		
Throughput, tons/hr	133	266	400	50	100	150
Utilization, hr/year	Operating cost,* $					
2000	19,450	23,975	24,375	1,775‡‡		
4000	33,150	43,550	53,750	3,550		
6000	47,100	62,525	81,125	5,325		
	Total cost,† $					
2000	84,970	90,495	94,985	59,925	82,550	108,825
4000	98,670	109,070	119,270	61,700	84,325	110,600
6000	112,620	128,045	147,645	63,475	102,375	112,375
	Cost/ton,‡ $					
2000	0.32	0.17	0.12	0.60	0.41	0.36
4000	0.18	0.10	0.07	0.31	0.21	0.18
6000	0.14	0.08	0.06	0.21	0.14	0.12
Plant construction§	232,000			210,000		
Amortization¶	65,520			27,300		
Source cost**	272,000			140,000	250,000	380,000
Yearly source expense††				27,850	53,475	79,750

*Operating cost covers labor, service, and maintenance.
†Total cost is the sum of operating cost, amortization, and yearly source expense.
‡Cost/ton is the quotient of total cost divided by tons/year.
§For maximum throughput in each case.
¶Over 10 years at 6%.
**For a 3-Mev Dynamitron, installed, and for ^{60}Co sufficient to provide 16 krads at each throughput rate.
††Cobalt replacement, interest, and amortization.
‡‡Operating cost is the same at all three throughput rates.

Legislation and Clearances of Irradiated Food

A survey of the situation in Europe was presented by Spaander.[65] The word *licensing* refers to every legal system under which the user of a radiation source is obliged to have official approval before using the source. In some countries, e.g., Norway, legislation is kept to a minimum because detailed legislation becomes out of date quickly. Belgium, Switzerland, and the USSR were cited as nations with too-detailed legislation. The Joint FAO/WHO/IAEA Expert Committee on the technical basis for legislation on irradiated foods (Rome, Apr. 21–28, 1964) recommended: (1) governments should establish legislation to control the production, importation, exportation, and distribution of irradiated foods; (2) requirements for establishing the safety for consumption of irradiated food should be laid down; (3) legal control should be based on the principle of a "positive list" of individually permitted foods and their radiation treatments; and (4) distribution of an irradiated food item should be permitted only after acceptance by the appropriate government agency of evidence that the food is wholesome. Wholesomeness is divided into toxicological safety, nutritional safety, and microbiological safety. National legislation should facilitate international cooperation. A license system for radiation sources is in force in 20 out of 25 European countries (Table 3), but specific regulations on food irradiation and on the importation and trade of irradiated food are in force in only 4.

In Belgium, according to Lafontaine,[66] a very restrictive attitude has been adopted because of the uncertain information about the possible effect of irradiated food on humans. Articles 64.1 (*b*) and (*c*) of Royal Decrees of Feb. 28, 1963, prohibit the addition of radioactive substances to foodstuffs and the treatment of foodstuffs by ionizing radiations. However, Articles 65 (*a*) and (*b*) provide for rescindment on the condition that the addition or treatment is for research purposes and that a license is obtained. The import, possession, or transport of food to which radioactive substances have been added or which has been treated with ionizing radiation is prohibited under Article 64.2.

In the United States, as reported by Josephson,[67] highly acceptable, wholesome, shelf-stable bacon, ham, pork, beef, chicken, and shrimp have been produced in the laboratory; and the shelf life of fish, wheat and wheat products, oranges, and potatoes has been extended. However, before commercial quantities are produced for the consumer market, approval is needed from the U. S. Food and Drug Administration. In addition, meat from mammals and birds needs approval from the U. S. Department of Agriculture. These agencies have approved irradiation for bacon, wheat, wheat products, and white potatoes. Collecting the data for successful petitions for clearance has proved to be time consuming and expensive, but this could be remedied by setting up clear-cut procedures in advance for evaluating data and by having the responsible agency advise and assist the petitioner during preparation of the petition.

Table 3 RADIATION LEGISLATION IN EUROPE

Country	License required for radioactive substances	License required for other radiation sources	Specific regulations on food irradiation	Specific regulations on importation or trade
Iceland	*	*	†	†
United Kingdom	*	*	‡	‡
Denmark	*	*	†	†
Sweden	*	*	†	†
Norway	*	*	†	†
Finland	*	*	†	†
Netherlands	*	§	‡	‡
Belgium	*	*	*	*
Luxembourg	‡	‡	‡	‡
France	*	*	†	†
Portugal	*	*	†	†
Spain	*	†	†	†
Federal Republic of Germany	*	*	*	*
Switzerland	*	*	*	*
Italy	*	†	*	*
Austria	§	§	†	†
Czechoslovakia	*	*	†	†
Poland	§	§	†	†
Hungary	†	†	†	†
Yugoslavia	†	†	†	†
Greece	†	†	†	†
Turkey	¶	†	†	†
Ukraine	*	*	†	†
USSR	*	*	†	†
Byelorussia	*	*	†	†

*Legislation in force.
†No data available.
‡Legislation in preparation.
§Legislation partly in force, partly in preparation.
¶Legislation exists, not in force.

A new procedure is being tried in Israel. Foa et al.[68] have two test cases before the Israel Ministry of Health with petitions based not on local work but on that published abroad. They are requesting that wholesomeness tests approved by other governments be accepted as sufficient proof of safety. Such a measure, if successful, would enable the small and developing countries to benefit most from irradiation of food since they could pass legislation without having to spend vast sums on testing the irradiated foodstuff. This procedure could also facilitate international trade agreements.

Goresline[69] set up standards for legislation to regulate all three categories of irradiation treatments — radappertization, radicidation, and radurization. Legislative controls for radappertization should ensure that adequate scientific data establish the destructive dose range for specific food. The necessary dose would then be a requirement for treating the food; proof of the accuracy of delivery of the dose would be demonstrated by data automati-

cally recorded during plant operation. Legislation on radicidation should require proof that the dose proposed does control the specific pathogen in the specific food. The regulations should spell out the specific treatment procedure and the safeguards to be used to prevent recontamination. Legislation on radurization should ensure that the treatment has been used to accomplish the intended results and not to cover up the inferiority of a product by reducing the microbial population and should guard against a possible assumption or claim that the product is pathogen free because of the treatment. All legislation on irradiation of food should be deliberately designed to facilitate international exchange of irradiated food. (F. E. McKinney)

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FDA Regulations for Irradiated Foods and Packaging Materials

By E. A. Garlock*

[*Editor's Note:* The recent reluctance of the Food and Drug Administration (FDA) to accept the data submitted in support of petitions for clearances of a number of irradiated foods has pointed out the advisability of including in *Isotopes and Radiation Technology* a discussion of what is expected. In this way organizations involved in food-irradiation experiments will be better able to anticipate the FDA requirements and can properly plan their evaluation *prior* to the start of their investigations. Unfortunately there has been a tremendous amount of wasted effort—brought about largely by the fact that this is a new area of endeavor and specifications had to be developed en route; many of these early specifications have had to be redone, thereby necessitating considerable additional (and often repetitive) work on the part of the petitioners. Hazleton Laboratories, Inc., who supplied the following article, has a contract with the U. S. Atomic Energy Commission's (AEC) Division of Isotopes Development (DID) for coordinating the petitions program.]

Abstract: The types of information required by the Food and Drug Administration to support petitions for clearance of irradiated foods are outlined. A set of guidelines is included.

The FDA has recently issued statements and guidelines designed to aid petitioners in complying with the regulations for the safe use of food additives, antibiotics, and drugs. These guidelines point out the type of data required in support of petitions and other documents requesting approval to market products regulated in interstate commerce. That FDA is becoming increasingly critical in their evaluation of safety and effectiveness data is apparent from a review of these guidelines. We have received requests for additional data as a result of FDA's review of our irradiated-food and packaging-materials petitions, and the need to fill these requests delays the review and approval of petitions. In order to reply to these requests, we must review data contained in contractors' reports and in the literature. In some cases experiments must be repeated and new studies initiated. A case in point is the recent request for experimental data to validate the statement that ionizing radiation does not induce detectable amounts of radioactivity in flexible packaging materials. Final action on the packaging-materials petitions has been deferred by FDA until the results of the studies on induced radioactivity are available.

According to the protocol included in the FDA guidelines, petitions on packaging materials are considered incomplete unless they contain the following details of the proposed food-packaging usage:

Individual foods or types of foods (specific examples) to be packaged

Maximum temperature of packaging

Maximum temperature and time of storage

Maximum temperature and time of cooking if food is to be subsequently processed in the package or container

Minimum size of package (weight of food) to be used

Area, mil thickness, and weight of film or other container such as a bottle or coating exposed to food in the minimum-size package

Extraction data to show the likely degree of migration to food of all components of the finished formulated packaging materials under the most severe conditions as well as under the usual conditions of proposed usage

Estimation of the maximum as well as average quantity of food additive to be expected in individual foods or types of foods and in the total daily diet of the consumer. (The basis for the estimation is to be described in detail, including any assumptions made and the arithmetical computations involved.)

This is just one of many examples of the detailed information which must now be supplied in support of food-additive petitions.

In the past we have had to rely on FDA guidelines that were written primarily to aid in the preparation of petitions on chemical additives. Obviously, many of the sections included in these guidelines are not directly applicable to irradiated foods, and this has resulted in unnecessary delays in the preparation and review of petitions. In an attempt to improve the situation, Hazleton Laboratories personnel have prepared and circulated suggestions* for AEC contractors in preparing reports on irradiated-food studies.

More important, however, is the recent FDA set of guidelines that includes the protocol for radiation and radiation sources used in treating or processing foods. A limited mailing of these guidelines has been made, and they are reproduced in the Sept. 19, 1966, issue of *Food Chemical News*. The following describes the information that must be included in irradiated food-additive petitions:

Protocol for Radiation and Radiation Sources

A. Identity:
1. Radiation and radiation source proposed: A. If an isotope, identify it, and describe the type of en-

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*See pages 140-141, this article.

capsulation used; B. If a machine source, describe it.

2. Engineering data providing complete information on the geometry of the radiating mechanism, including the speed of movement by the radiating head, the radioisotope, and the number of curies in the source used (if isotopes), the voltage and amperage used (if machine), and the total time of exposure. A statement of the radiation flux calculated therefrom should be included.
3. Data providing complete information on the means to be used to control the exposure: A. In the case of machines, data showing the sensitivity of the means of controlling the power sources, and the way in which recorders are coupled to produce records of the operating voltages and amperages; B. In the case of isotopes and machines, describe the nature of the dosimeter and the frequency of the dosimetry, and furnish data showing the dosimetry and phantoms to be used, including those which show that the dosimetry readings will adequately reflect the dose absorbed by the food during exposure.

B. Use:
This section should include information on the dose ranges to be used and the purpose for which the radiation is proposed, together with all directions, recommendations, and suggestions regarding the proposed use. This section should also include specimens of labeling to be used on the radiation sources and any labeling to be used on the treated food.

C. Technical Effect:
1. Experimental data should be submitted showing that the radiation dose proposed accomplishes the intended technical effect and does not exceed the amount reasonably required to accomplish the intended effect.
2. Information should be included showing that there is no induced radioactivity in the food or packaging material. This can include a theoretical discussion but some actual experimentation data should be provided to support the theory advanced. If different radiation fluxes are to be used, those materials subject to the highest contemplated flux and dose should be selected for testing for induced radioactivity. A description of the methodology and the sensitivity of the methods used to establish the absence of induced radioactivity in the treated foods or packaging material should be furnished.
3. Data should be provided to establish the extent to which the nutritive composition of the irradiated food has been altered by exposure of the food to the highest dose and under the highest flux expected. If any such changes are felt to be insignificant, these should be so designated and the reasons detailed for their claimed insignificance. The methodology employed should be fully described.
4. Data should be provided to establish that the irradiated food has not been significantly altered in organoleptic or other physical characteristics so as to render the material otherwise unfit for food. Where a fresh food is ordinarily stored for a period of time at particular temperatures, data should be included reflecting the results when the irradiated food is appropriately stored and/or shipped.

These guidelines have evolved in the course of negotiating and resolving various problems in connection with the preparation and review of petitions for the safe use of radiation. The importance of these guidelines is amplified by the fact that they have been formally issued by the regulating agency. In addition to these petition requirements, the Acting Associate Commissioner for Compliance for FDA, J. K. Kirk, called attention before a recent session of the Joint Committee on Atomic Energy to the fact that the statute specifically directs in part that "no such regulations shall issue if a fair evaluation of the data ... (A) fails to establish that the proposed use of the food additive under the conditions of use to be specified in the regulation will be safe. ... Provided, that no additive shall be deemed to be safe if it is found to induce cancer when ingested by man or animals ..., or (B) shows that the proposed use of the additive would promote deception of the consumer in violation of this Act or would otherwise result in adulteration or misbranding of food within the meaning of this Act."

Kirk also referred to the published amendments to several of the irradiated-food regulations requiring such labeling statements as "PROCESSED (OR TREATED) BY IONIZING RADIATION" and in the case of bulk shipments, the words "DO NOT IRRADIATE AGAIN." Comments in response to these labeling statements are being evaluated by the FDA to determine if a hearing should be granted to resolve the issues.

In summary, the two most important deficiencies in meeting current FDA petition requirements are furnishing (1) raw experimental data to support calculated values and (2) data on the effectiveness of radiation for preserving perishable foods during commercial handling. For example, FDA will not complete the review of an irradiated-fruit petition unless raw data, such as actual numbers of fruit used in the experiment, numbers of spoiled fruit, and numbers of microorganisms, are included for purposes of calculating relative effects. FDA is also reluctant to act favorably on a petition that does not include effectiveness data on commercial shipping, storage, and distribution studies. The effectiveness of irradiation of commercial quantities of food must be included in each petition. To collect commercial shipping and storage data on oranges, nine master cartons, each containing eight 5-lb bags of oranges that had been separated into three groups and respectively treated at 0, 150, and 300 krads of gamma radiation, were shipped from Jacksonville, Fla., via REA Express without refrigeration to Washington, D. C. Upon arrival the cartons were placed in storage at 35 and 62 °F at Hazleton Laboratories, and after 9 and 39 days' storage, fruits were evaluated for weight loss, spoilage, and peel injury. Juice from the fruit was analyzed for solids, *p*H, percentage of titratable acidity, and reduced ascorbic acid. For organoleptic evaluations the juice was served to a taste panel of 10 judges and rated for color, consistency, and flavor. The results of this study have been reported and submitted to FDA as a supplement to the irradiated-orange petition. This is the kind of

data which must be included in all food-additive petitions, and investigators should plan to initiate these shipping, storage, and acceptability studies as soon as they can be scheduled. It should be remembered, however, that no irradiated food can be shipped in interstate commerce unless a regulation has been issued providing for the safe use of that food item or unless it is an experimental shipment to a responsible investigator and clearly marked with the statement "CAUTION: CONTAINS A NEW FOOD ADDITIVE FOR INVESTIGATIONAL USE ONLY IN LABORATORY RESEARCH ANIMALS OR FOR TESTS IN VITRO. NOT FOR USE IN HUMANS" (F. R. 4891, Mar. 24, 1966 — Fig. 1).

Requests must be submitted to the FDA for permission to conduct consumer type acceptability studies on irradiated foods. Each request must include a description of the experimental design, data showing that the product is wholesome, and the precautions that will be taken to ensure that none of the experimental food will enter retail marketing channels. Competent investigators must supervise the distri-

(§§ 121.74, 121.75)

*Amendment published in *Federal Register*, Mar. 24, 1966; 31 F.R. 4891.

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§ 121.74 Procedure for amending and repealing tolerances or exemptions from tolerances.

(a) The Commissioner or any interested person may propose the issuance of a regulation amending or repealing a regulation pertaining to a food additive or granting or repealing an exemption for such additive. Such a proposal by an interested person shall be in writing. If such proposal by an interested person furnishes reasonable grounds therefor, the Commissioner will publish a notice announcing the proposal. Proposals initiated by the Commissioner will likewise be published. Following such publication, the proceedings shall be the same as prescribed by section 409 of the act and the regulations in this part for the promulgation of a regulation.

(b) "Reasonable grounds" shall include an explanation showing wherein the person has a substantial interest in such regulation and an assertion of facts (supported by data if available) showing that new information exists with respect to the food additive or that new uses have been developed or old uses abandoned, that new data are available as to toxicity of the chemical, or that experience with the existing regulation or exemption may justify its amendment or repeal. New data should be furnished in the form specified in § 121.51 for submitting petitions.

***§ 121.75 Exemption for investigational use and procedure for obtaining authorization to market edible products from experimental animals.**

A food additive, including one that is a new drug or antibiotic, or foods containing such a food additive, intended for investigational use by qualified experts shall be exempt from the requirements of section 409 of the act under the following conditions:

(a) If intended for investigational use in vitro or with laboratory research animals, it bears a label which states prominently, in addition to other information required by the act:

Caution—Contains a new food additive (or new drug, or antibiotic) for investigational use only in laboratory research animals, or for tests in vitro. Not for use in humans.

(b) If intended for clinical investigational use in animals other than laboratory research animals, and the edible products of the animals are to be marketed as food, permission for the marketing of the edible products as food has been requested by the sponsor, and authorization has been granted by the Food and Drug Administration in accordance with §130.3a of this chapter, or by the Department of Agriculture in accordance with § 309.20 of Title 9, Code of Federal Regulations, and it bears a label which states prominently, in addition to other information required by the act, the following:

Caution—Contains a new food additive (or new drug, or antibiotic) for use only in investigational animals in clinical trials. Not for use in humans.

Edible products of investigational animals are not to be used for food unless authorization has been granted by the U.S. Food and Drug Administration or by the U.S. Department of Agriculture.

Figure 1

bution, record keeping, and disposal of the irradiated products.

If contractors and investigators adhere to these guidelines in planning experiments and reporting the results, chances of expediting the clearance of irradiated foods will be greatly increased. (PSB)

Guidelines for AEC Food-Irradiation Contractors

By E. A. Garlock and J. M. Barnes*

I. General Requirements for Preparation of Food Additive Petition

The following guidelines are based upon recent experience and upon regulations stated in the Food, Drug, and Cosmetic Act (amended), Section 409, to familiarize contractors and sponsoring agencies with the scope and requirements necessary to bring research programs on a food item to a position suitable for petition purposes.

A. The objectives of the projects should be specifically stated to evaluate safe use of the food additive (ionizing energy) and define conditions for irradiation with the following parameters in mind:
   1. Radiation source employed and methods used to insure uniform dose control.
   2. Establishment of a dose range which will provide optimum beneficial effects under anticipated commercial conditions.
   3. Type of dosage monitoring that is compatible with AEC licensing requirements and proposed commercial radiation systems.

B. The potential benefits accruing through use of radiation should be defined under the proposed conditions:
   1. Control of spoilage organisms and extension of market life.
   2. Control of contaminant organisms or their by-products.
   3. Improvement of general quality and appearance of the food item.

C. Appropriate studies should be conducted on the indirect effects of radiation such as induced radioactivity or effects on radiosensitive vitamins.

D. Experimental protocol should be clearly and succinctly defined in conformance with FDA guidelines.

E. Those concerned with food irradiation programs should be aware that the FDA requires a description of practicable analytical methods for control and regulatory purposes, description and design of irradiation facility including maximum product throughput and practicable monitoring and recording to assure acceptable dosage control.

II. Format of Research Reports in Support of Petitions to Be Submitted to FDA

The following suggestions are derived from experience of Hazleton Laboratories personnel with FDA in filing and following up Food Additive Petitions under the provisions of the Food, Drug, and Cosmetic Act (amended), Section 409.

A. Research reports should contain tabular or graphic presentations of results, accompanied by appropriate statistical analysis of significant effects. The type of statistical analysis should be tailored to the data presented. It is impractical for outside groups to dictate the type of analysis to be used for each study because of the wide variety of studies.

B. Research reports should contain definitive conclusions on the research in such a manner that they can be expeditiously incorporated into FDA petitions.

C. Animal feeding studies should be reported in full, giving the experimental design, general physical appearance, behavior, signs of toxic or pharmacologic effects, and gross and individual microscopic necropsy findings.
   1. Subacute Oral Administration — Male and Female Dogs, 90-Day Observation Period: Tables should be furnished showing individual weekly body weights, and compound consumption; individual hematological, biochemical and urinary values; and individual organ weights and organ/body weight ratios. Statistical evaluation of the data should be performed where appropriate.
   2. Subacute Dietary Administration — Male and Female Rats, 90-Day Observation Period: Statistical evaluation of body weights, food consumption, survival, organ weights, organ/body weight ratios, and hematological values should be performed. Tables should be furnished showing mean weekly body weights, weight ranges, food consumption, and survival data; individual hematological and biochemical values; results of urine analyses; and mean terminal body weights, organ weights, and organ/body weight ratios. Mean weekly body weights and food and compound consumption should be presented graphically.
   3. Chronic Dietary Administration — Male and Female Dogs, Two-Year Observation Period: Organ weights, organ/body weight ratios, and gross and individual microscopic necropsy findings should be reported. Statistical evaluation of body weight, food consumption, survival, organ weights, organ/body weight ratios, and clinical values should be performed where indicated. Tables should be furnished showing (a) individual body weights at initiation, the first, second, and fourth weeks and for every fourth week thereafter; (b) individual food and compound consumption for the first, second, and fourth weeks and for every fourth week thereafter; (c) individual clinical values; and (d) individual organ weights and organ/body weight ratios.
   4. Chronic Dietary Administration — Male and Female Rats, Two-Year Observation Period: Organ weights, organ/body weight ratios, and gross and microscopic necropsy findings should be reported. Statistical evaluation of body weight gains, food consumption, survival, organ weights, organ/body weight ratios, and clinical values should be performed where indicated. Tables should be furnished showing (a) mean weekly body weights, weight ranges, food consumption, and survival data; (b) individual clinical values; and (c) mean organ weights and organ/body weight ratios. Mean weekly body weights, food consumption, and compound consumption should be presented graphically.

III. Effectiveness Reports

These reports are concerned with the effects that the food additive (ionizing radiation) is intended to have and quantity of such additive necessary to achieve

*Hazleton Laboratories, Inc.

the intended effect. The FDA has requested that original data be submitted with petitions. In these studies, as well as in safety and wholesomeness studies, statistical evaluation of the data is recommended. Tests of significance for any treatment effects or interactions should be presented in tabular form, and also be presented in the discussion and/or summary sections of reports. Specific suggestions for reporting results of certain studies pertaining to effectiveness are presented below:

A. Product Quality Research
   1. Organoleptic Evaluations
      The actual numbers of participants on taste panels should be presented. The actual distribution of judgments among various quality attributes and among treatments should be shown.
   2. Appearance and Textural Studies
      Color photographs showing appearance of product as affected by treatment should be presented. Where appropriate, results of quantitative determinations of pigment content of various lots should be furnished. Data from shear-press or other pressure gauge determinations of texture or firmness should be presented in raw form.

B. Shipping and Storage/Shelf-life Research
   Actual number of days of shelf-life extension, accompanied by presentation of quantity of food item used in the study, is desirable. Laboratory-scale shipping and storage studies are of little value for FDA review purposes. Simulated shipping and marketing tests are of value only in evaluating test parameters for later tests under actual conditions. Studies involving food items in wholesale or retail containers in sufficient numbers under actual commercial conditions for meaningful statistical evaluation should be conducted.
   Reports on storage decay control should contain data on actual numbers of fruit or other food items showing decay relative to total number in each treatment. Actual microbial plate counts and statistical evaluation of results should be presented.

The above guidelines are for general information to contractors involved in the AEC irradiated foods program.

Section

IV

Isotopes and Radiation Technology

Process Radiation Development

Chemonuclear and Radiation Chemical Process Research and Development

By Meyer Steinberg*

Abstract: The guiding principles and some of the latest results in the development of nuclear energy for synthesis and production of industrial and utility chemicals are presented. This work indicates that fissiochemonuclear processes for fixation of nitrogen, synthesis of ozone, and production of synthesis gas (carbon monoxide and hydrogen) are sufficiently interesting, from a long-term national-goals point of view, to provide ample incentive for continued orderly development of the science and technology of this field. Radiation-induced chemical processes using isotopic and machine sources for chain-reaction products will continue attracting further industrial development efforts.

The development of chemonuclear processing, defined as the use of nuclear reactors and large radiation sources for industrial chemical production, has its place among the many other peaceful applications of nuclear energy being pursued under the direction of the U. S. Atomic Energy Act. Whereas the development of nuclear energy for electric power, ship propulsion, and special terrestrial and space applications has already, more or less, come into practical use, the development for chemical processing is still very much in the research and early developmental stage. Although the size of budget involved does not yet reflect it, the chemonuclear program has a potential similar to that of the development of nuclear reactors for desalting seawater and of nuclear explosives for commercial purposes. The underlying problem for all these peacetime applications of atomic energy is the same, that is, overcoming the economic barrier of the system. It must be emphasized that the broad definition of chemonuclear processing includes not only the use of the direct chemical effects of the high-energy radiation produced by the nuclear fission reaction but also of the available thermal, electrical, and photolytic energies for promoting industrial chemical reactions. In fact, it appears that a solution to the economic problem may lie in utilizing a combination of these effects in multicycle and multipurpose systems. Since most of the nuclear energy first makes its appearance in the form of the kinetic energy of the recoiling fission fragments, advances in the direct use of the fission-fragment recoil energy might prove economically interesting.

A few facts and figures may put the program into perspective. The current[1] U. S. energy consumption is approximately 1.7 million Mw(t)-years, of which some 15%, or 250 thousand Mw(t)-years, is utilized by the chemical industry.[2] These demands are expected to increase sevenfold by the end of the century. The chemical industry, which will be contributing to the predicted rising energy consumption during this period, will have the added burden of competing for fossil hydrocarbon resources as special raw materials for chemical products. The nation and the chemical industry must be mindful of preserving the natural carbon–hydrogen bond reserves and, when necessary, provide means for re-forming them.

One increasing demand for the destruction of natural C–H bonds is in the production of hydrogen needed for fixation of nitrogen as fertilizer, so vital to increased world food requirements. It is estimated that the present U. S. capacity of 8.5 million tons of ammonia per year accounts for about 2.2% of the total natural-gas consumption. The ammonia production capacity is growing at a rate of greater than 20% per year,[3] compared to the normal rise of 6% per year for natural-gas supplies. Thus in a few years — by 1968–1970 — when the ammonia production capacity is expected to rise to over 18 million

*Brookhaven National Laboratory, Upton, N. Y. Paper presented at American Nuclear Society Topical Meeting on Large-Scale Production and Application of Radioisotopes, Augusta, Ga., March 20–23, 1966. The author acknowledges the valuable assistance and advice of B. Manowitz, Head of the Radiation Division, and W. D. Tucker, J. W. Sutherland, M. Beller, J. Sears, G. Farber, E. Cote, L. E. Kukacka, P. Colombo, J. Fontana, and J. Prozansky of the Nuclear Engineering Department in developing the material for this paper.

tons per year, the demand may rise to about 7% of the total natural-gas consumption. If this rate of increase continues, it is probable that other sources of hydrogen will be turned to, including liquid and solid fuels. The chemonuclear program offers the potential, in this one area alone, of relieving this demand. Research has indicated that nitrogen can be fixed directly by using high-energy fission-fragment radiation and indirectly by producing hydrogen through electrochemical decomposition of water.

Fissiochemonuclear research investigates the use of fission-fragment energy for promoting endothermic, nonchain, chemical reactions, which are typically low-G-value reactions. In a sense a fissiochemical reaction is a direct conversion of nuclear to chemical energy. A brief review of some guiding principles[4-9] and highlights of the latest results are presented here under the headings Fission-Fragment and Radiation Chemistry; Chemonuclear Source and Fuel Development; In-Pile Loop Experiments; Economic Evaluation; Contamination Problems; and Radiation Chemical Processing.

The area where each type of high-energy radiation system generally applies is shown graphically in Fig. 1. This generalized figure indicates the reactor

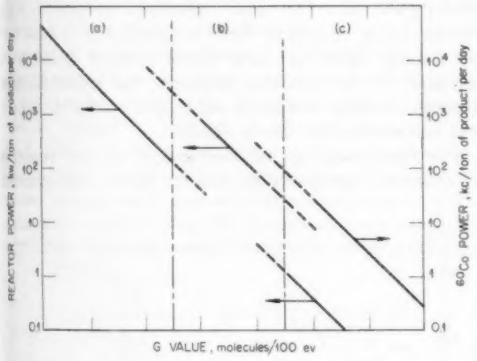

Fig. 1 Power requirements as a function of G value for chemonuclear reactions; average product molecular weight of 50 assumed. (a) *Nonchain reactions, endothermic, with fission-fragment reactor, e = 50%;* (b, c) *chain reactions, exothermic, with* (b) *(n,γ) reactor, e = 2.5%, and* (c) ^{60}*Co irradiator and machine, e = 50%.*

power per unit of chemical production rate as a function of G value and is based on the expressions

$$E = 1216/Ge_0M \quad (1)$$

$$P = 101.5T/Ge_0M \quad (2)$$

$$C = 6.95 \times 10^6 T/Ge_0M \quad (3)$$

where G = yield of chemical product, molecules/100 ev
E = energy requirement per unit of chemical product, kw(t)-hr/lb
e_0 = radiation-energy-deposition efficiency or fraction of fission energy (based on 200 Mev/fission) deposited directly as high-energy radiation in the reaction system
M = molecular weight of product or repeating unit (in case of polymer product)
P = thermal power level of chemonuclear reactor or high-energy machine, Mw(t)
T = production rate of chemical, tons/day
C = power level of ^{60}Co source, kilocuries

For large-scale chemical production rates of tens to hundreds of tons per day, a fission-fragment reactor is the only means for reaching the required power levels of tens to thousands of megawatts of radiation power for reactions having G values lower than 30, i.e., for endothermic, nonchain, reactions. An (n,γ) reactor can be considered for G values ranging from tens to thousands, while isotopic and machine sources can be considered only for G values in the range of thousands to hundreds of thousands, i.e., for exothermic, chain, reactions.

The systems that have recently attracted the greatest interest for fission-fragment studies are listed in Table 1. The experimental and theoretical maximum G values and the thermal efficiency, given for each reaction, are defined by

$$G_{max.} = 2.307 \times 10^3/\Delta H \quad (4)$$

$$T.E. = G_{exp}\,\Delta H/23.07 \quad (5)$$

where $G_{max.}$ = maximum G value, molecules/100 ev
G_{exp} = experimental G value, molecules/100 ev
ΔH = heat of reaction, kcal/mole of product
T.E. = thermal efficiency of radiation-induced reaction, %

The G values listed were selected from among the highest reported in the literature for any type of high-energy radiation.

Fission-Fragment and Radiation Chemistry

FIXATION OF NITROGEN

An extensive series of in-pile capsule irradiations of nitrogen–oxygen gas mixtures has been made using a fission-fragment source and yield–dose–product buildup curves obtained for the pressure range 3 to 68 atm and for nitrogen concentrations from 30 to 90%. In addition, ^{60}Co gamma and reactor radiation $(n + p + \gamma)$ have also been used as radia-

Table 1 FISSION-FRAGMENT CHEMONUCLEAR REACTIONS

Reaction* (product underlined)	Heat of reaction at 25°C, kcal/mole	$G_{max.}$	G_{exp}	Energy, kw(t)-hr/lb	Thermal efficiency, %
$\frac{1}{2}N_2(g) + O_2(g) = \underline{NO_2}(g)$	+8.1	284	6	4.4	2.1
$\frac{3}{2}O_2(g) = \underline{O_3}(g)$	+34.0	68.1	15	1.7	22.0
$CO_2(g) = \underline{CO}(g) + \frac{1}{2}O_2(g)$	+67.6	34.1	10	4.3	29.4
$H_2O(g) = \underline{H_2}(g) + \frac{1}{2}O_2(g)$	+57.9	39.9	6	101.5	15.0
$2H_2O(l) = \underline{H_2O_2}(l) + H_2(g)$	+91.4	25.2	1.7	21.0	6.8
$2NH_3(l) = \underline{N_2H_4}(l) + H_2(g)$	+48.0	48.1	2	19.0	4.2
$\frac{1}{2}N_2(g) + CH_4(g) = \underline{HCN}(g) + \frac{3}{2}H_2(g)$	+49.0	47.2	2	22.5	4.2

*Here (g) and (l) refer to gaseous and liquid states.

tion sources under similar conditions. The significant findings can be summarized as follows:

(1) For a 77.3% N_2 + 22.7% O_2 gas mixture (simulating the composition of air), the total initial G value for NO_2 formation, $G_i(NO_2)_T$,* increases from 0.76 to 1.21 over the pressure range 4.8 to 64.8 atm at 30°C. At the same time the $G_i(N_2O)$ value decreases from 0.69 to 0.27.

(2) The $G_i(NO_2)$ value does not vary appreciably up to 90% N_2 but decreases markedly above this concentration.

(3) The $G_i(NO_2)_T$ value varies only between 0.7 and 1.2 as the radiation source is changed from ^{60}Co gamma to reactor $(n + p + \gamma)$ to fission fragment $(f, f + n + p + \gamma)$ for a 77.3% N_2 + 22.7% O_2 gas over the pressure range 5 to 63 atm at 30°C. The $G_i(N_2O)$ value varies between 0.3 and 0.7. Only for reactor radiation at 66 atm does the $G_i(N_2O)$ value decrease to the low value of 0.05.

(4) Relatively high concentrations of nitrogen oxides can be built up in a N_2-O_2 gas mixture in a radiation field. At 5 atm pressure and 30°C, more than 90% of the oxygen present can be converted to nitrogen oxides to give a steady-state composition of 7.5% N_2O_4, 6.3% NO_2, and 6.9% N_2O.

(5) When high doses ($>75 \times 10^{22}$ ev) of fission-fragment radiation are given to the N_2-O_2 gas mixture at 65 atm, sufficient NO_2 is made that liquid N_2O_4 condenses in the capsule during the irradiation. There is an apparent increase in $G(NO_2)$ while the N_2O concentration remains at steady state. The data obtained are scattered due to the difficulty in carrying out experiments in this high NO_2 concentration and dose range because of the corrosive effect on the radiation source. Several experiments with corrosion-resistant platinum-coated foils (Fig. 2) have, however, confirmed the high NO_2 concentrations. Although it is apparent that more precise experimental results are needed, it is interesting that, for doses of the order of 100×10^{22} ev, the differential or incremental $G(NO_2)$ value is in the range 3 to 9. At the highest dose given to the gas mixture, approximately 2% of the oxygen remains in the system. Much interesting speculation surrounds this observation, centering mainly around the importance of the liquid N_2O_4 phase and its possible reaction with N_2 and/or N_2O in producing NO and NO_2. To take advantage of this effect in a process system, it would be necessary only to adjust the reactor-feed gas and liquid compositions to the range where the highest incremental G value is obtained and the flow rate to obtain the corresponding high concentration increase.

In a separate series of experiments, it has been established that N_2 gas dissolved in liquid N_2O_4 reacts in a radiation field to yield NO. These observations open up new paths toward fixation of nitrogen by a radiation process, and much further experimentation and basic study are needed to establish and understand these results.

For obtaining an understanding of the reaction mechanism, several experiments have been carried

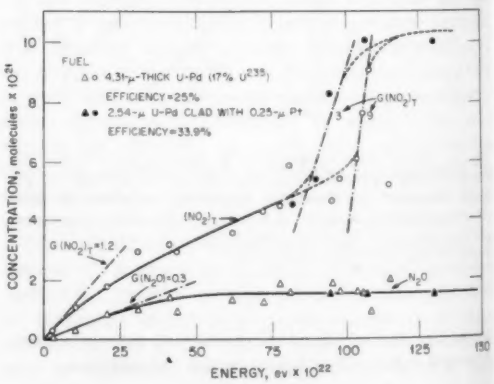

Fig. 2 Experimentally determined yield–dose curves for fission-fragment and reactor irradiation $(n + p + \gamma)$ of a 77.3% N_2 + 22.7% O_2 gas. Flux, (0.8 to 1.1) × 10^{13} n/(cm²)(sec); vessel volume, approximately 32 ml; ^{235}U, approximately 10 mg; pressure, 65 atm; temperature, approximately 35°C.

*The subscript T indicates total NO_2, i.e., N_2O_4 plus NO_2, and the subscript i refers to initial G values.

out to establish a G value for monatomic nitrogen formation in N_2 gas.[7,10,11] These have been based mainly on studies of the $^{14}N_2-^{15}N_2$ exchange in a radiation field. The G(N) value may possibly be useful in establishing an upper G limit for nitrogen fixation. However, the system has been found extremely complicated, possibly due to formation of ionized and excited N_4 species. A lower G(N) limit[11] of 2.5 seems to be established; however, it may be[7] as high as 11.5.

Other interesting possibilities include the effect of radiation on the high-temperature (above 1000°C) reaction[12] between N_2 and O_2 and the reaction of nitrogen dissolved in water[13] to form nitrates directly in a condensed aqueous phase.

It should be mentioned that two unique features of the high-energy-radiation fixation of nitrogen are that it is the only known process that can convert over 20% of the nitrogen in an air mixture to higher oxides of nitrogen in an one reaction step and that this can take place at a bulk temperature of 25°C. The problem remaining is to establish a higher yield based on radiation energy absorbed in the system.

OZONE SYNTHESIS

The relatively high G values and concentrations for ozone formation in liquid ozone[14] prompted initiating a study of the radiation and fission-fragment chemistry in the gas phase at temperatures where a process could be developed without excessively high refrigeration costs, i.e., at temperatures[15] down to -78°C. With ^{60}Co gamma irradiation of pure oxygen at 1 atm and 12°C in Pyrex and quartz vessels,[16] the highest $G_i(O_3)$ value obtained was 5.6, and the ozone steady-state concentration dependence on dose rate was marked (Fig. 3). Concentrations as high as 0.2% have been obtained. Extrapolations at dose rates

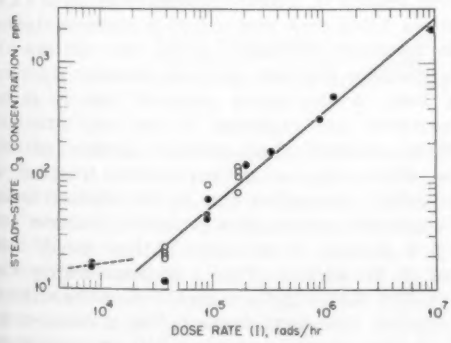

Fig. 3 Steady-state ozone concentrations as a function of dose rate in the gamma irradiation of pure oxygen in Pyrex and quartz at 12°C and 300 to 700 mm Hg pressure. The ozone concentration is proportional to $I^{0.83\pm0.06}$.

equivalent to those obtained in corona-discharge ozonizers give concentrations of the same order of magnitude (~8%). The 0.8-power intensity dependence on steady state is in disagreement with the postulated ion-chain decomposition back reaction.

An initial fission-fragment irradiation experiment at a temperature of approximately -78°C in the gas phase has indicated a G value for O_3 formation of at least 2.8. This result is the first known reported formation of O_3 using fission-fragment energy and will be pursued with interest.

In the irradiation of flowing N_2-O_2 gas at elevated pressures in a ^{60}Co gamma field, O_3 formation was noted at high flow rates without the formation of NO_2. Figure 4 shows the variation of O_3 and NO_2 concentrations as the flow rate is decreased for a 79% N_2 - 21%

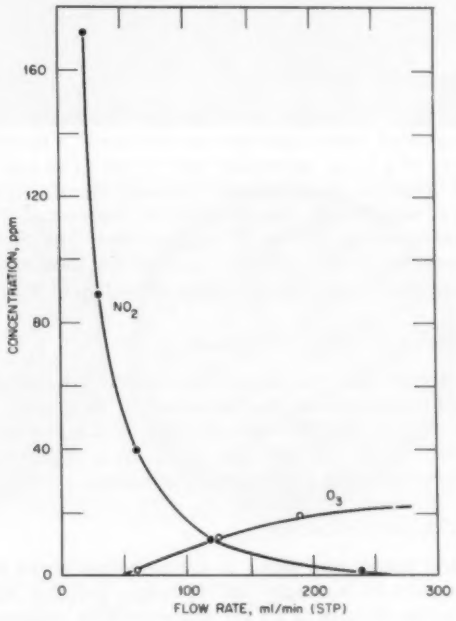

Fig. 4 Effect of decreasing flow rate on O_3 or NO_2 concentration in ^{60}Co irradiation of a flowing stream of 79% N_2 + 21% O_2. Conditions: 68 atm, 25°C, 673,000 rads/hr, $N_2O \leq$ 100 ppm.

O_2 mixture at 68 atm. The ozone concentration goes through a maximum and then decreases to zero, at which time NO_2 begins to appear in significant amount. A maximum O_3 concentration of 22 ppm and a $G(O_3)$ value of 4.3 are calculated from these data. The maximum $G(NO_2)$ value found under these conditions was 1.8. These results are significant for applications where dilute O_3 concentrations in an air stream can be used, as shown below.

CARBON DIOXIDE DECOMPOSITION

In the past, high G values for CO formation from gaseous CO_2 decomposition in a radiation field required the presence of NO_2 as an inhibitor to prevent back reactions.[17] Some recent experiments[18] using fission fragments in a flowing gaseous CO_2 system indicate G(CO) values from 13 to 20 and corresponding stoichiometric $G(O_2)$ values of 7 to 10, which require no NO_2 inhibitor for freshly prepared reaction vessels. Increasing the temperature from 200 to 400°C decreased the G values, but increasing the pressure from 20 to 27 atm tended to increase the yield. Concentrations of CO as high as 1% were obtained. Previously decomposition of CO_2 in a gamma-radiation field under static conditions was found[19] to yield a G(CO) value of 4.5. The reason for these higher results with fission-fragment radiation may be due to linear energy transfer (LET) or to a track effect.

HYDRAZINE SYNTHESIS

Latest information on low-dose ^{60}Co gamma radiolysis of liquid ammonia has indicated[20] a $G_f(N_2H_4)$ value of 2.0, in agreement with yields of as high as 1.9 found in some fission-fragment experiments.[21] Loop experiments have indicated[22] hydrazine concentrations as high as 1%. Higher dose ^{60}Co gamma irradiation gave a $G(N_2H_4)$ value of 0.2, indicating a track effect favoring a biradical formation of N_2H_4.

HYDROGEN CYANIDE SYNTHESIS

Another approach to nitrogen fixation has recently been undertaken in the formation of HCN from N_2 and CH_4. A G(HCN) value as high as 2.0 was measured[23] up to 14 atm and 600°C in a flowing-gas experiment using fission-fragment energy.

WATER DECOMPOSITION

It is known that water in the condensed phase decomposes to hydrogen and hydrogen peroxide with an initial G value of 1.7 for heavy-particle radiation. There is some evidence[24,25] that water in the gaseous phase decomposes more efficiently, and a $G(H_2)$ value as high as 6 has been reported. As a potential chemonuclear process, more studies are indicated for the fission-fragment decomposition of steam at elevated temperatures and pressures.

Chemonuclear Source and Fuel Development

The operation of a fissiochemonuclear reactor depends on the development of a workable fission-fragment source and reactor fuel element. Theory and experiment have indicated that metallic, electrically conducting, elements would be more radiation stable to fission-fragment damage than ceramic, electrically insulating, elements. The theory is based on an electron-cloud mode of energy dissipation of fission-fragment energy.[26] In ceramic elements, where there is a resistance to carrying off the electronic energy, damage occurs through formation of a thermal spike in the fission-fragment track. This effect has been observed in fission experiments in which uranium-containing glass fibers sintered and fused while ultrathin uranium–palladium alloy foils remained intact. At BNL, effort has been concentrated[27] on developing a 20 wt.% ^{235}U–80 wt.% palladium foil in thicknesses down to 2.5 μ (0.1 mil). One BNL standard source of 4 μ thickness has been used in the capsule irradiation studies, yielding a fission-fragment energy-deposition efficiency of 25%. Measurement of energy deposition has been based on the nitrous oxide gas dosimeter specifically calibrated and standardized in a gamma field.[28] The average range of the fission fragments in this foil is 5.6 μ, so that a 2.5-μ-thick foil yields an efficiency of 38%. The alloy, a solid solution with a melting point of approximately 1400°C, has been subjected to a burnup of 25% of the ^{235}U content in reactor fluxes of 5×10^{14} n/(cm^2)(sec) in pure helium, nitrogen, and oxygen atmospheres with few visible signs of damage. However, in a highly concentrated NO_2 atmosphere, bare uranium–palladium foil undergoes severe radiation damage and corrosion. This has been noted also in high-dose irradiation in air, where appreciable NO_2 concentrations are formed, and has been attributed to a synergistic oxidation and radiation damage effect due to surface formation of a nonconducting UO_2 coating. The problem was solved by coating the uranium–palladium foil with an extremely thin layer of platinum. This was done by applying a thicker layer of platinum to the slab and rolling the entire assembly on a Sendzimir mill. Figure 5 shows a cross section of a 2000× photomicrograph of a 2.5-μ foil—a 2.0-μ core with a 0.25-μ platinum cladding. The measured efficiency of the clad foil was 35%. The cladding may also serve the purpose of preventing loss of fissionable material due to surface evaporation and spallation. It has been estimated[29] that an uncoated clean uranium surface will lose nine atoms of uranium per fission fragment. An interesting observation also is that surface damage will probably occur when a fission fragment exits from a surface at an angle of less than 5° to the plane of the surface. Thus a platinum coating of not more than 0.35 μ (5.6 μ × tan 5°) should be sufficient to prevent long-term damage. The platinum-coated foil is being adopted as another BNL standard source for capsule experiments.

Some effort[30] has been going into the development of UO_2-containing nickel cermet fibers, which may have some promise for a random-array element.

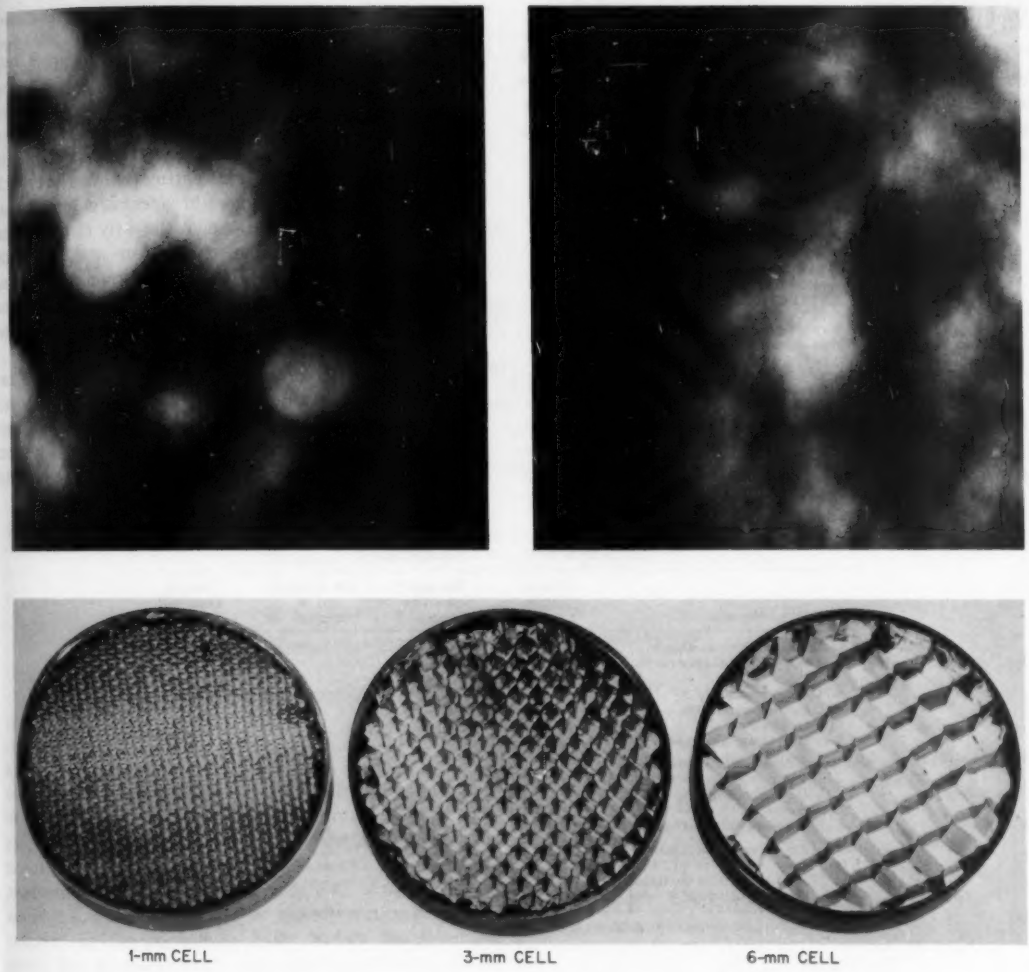

Fig. 5 Fissiochemonuclear fuel foil: 2.0-μ-thick core of 20% uranium–80% palladium, 0.25-μ-thick cladding of platinum. Above, micro cross section, 2000×; below, 5-mm-diameter honeycomb assemblies.

For thermal and fluid dynamic stability, the 2.5-μ fuel foil has been fabricated by Hexcel Products, Inc., of Berkeley, Calif., into a honeycomb assembly, using a diffusion-bonding technique. Honeycombs of three different cell sizes are also shown in Fig. 5. Each cell size may be used for a range of gas conditions, fixed by the maximum gas-phase range, so that a fission fragment does not reenter an adjacent foil member. These honeycomb assemblies have been subjected to flow rates of up to 100 ft/sec at 540°C for 1000 hr with no sign of mechanical failure. Quantity lengths of this element are being fabricated for the BNL in-pile loop experiment.

If construction of a critical reactor assembly of 2.5-μ foil turns out to be too much to ask for, foil elements may be mounted on structurally strong backing material in which half the fission-fragment energy may be lost. However, since the coating can be rolled even thinner, it may be reasonable to expect practical efficiencies approaching 35 to 40%. Reactor physics calculations have indicated the feasibility of attaining critical core assemblies with these honeycomb configurations for foil materials containing palladium, nickel, and aluminum. Uranium–aluminum foil on a sintered aluminum backing may be practical for an ozone-producing reactor.

In-Pile Loop Experiments

The Nuclear Engineering Department at BNL is constructing a versatile Chemonuclear In-Pile Research Loop (B-CIRL) to be placed in the Brookhaven Graphite Reactor. The design characteristics are

Gases to be handled	N_2O, N_2, O_2, NO_2, He, CO_2, CH_4, NH_3
Gas pressure range	0 to 1000 psig (1 to 69.1 atm)
Temperature range	−30 to +1000°F (−34.4 to +538°C)
Gas velocity	3 to 30 ft/sec (91.5 to 915 cm/sec)
Maximum volumetric flow rate	170 cu ft/min at 1 atm and 60°F (4550 liters/min at 1 atm and 0°C)
Fuel zone dimensions	1.5 in. in inside diameter by 5 ft long (3.8 cm in inside diameter by 1.53 m long)
Maximum flux	2×10^{12} n/(cm²)(sec)
Maximum fission power level	5000 watts (~50 g of ^{235}U)

The purpose of the loop is to (1) study the fission-fragment chemistry of a variety of gases in a flowing-gas system, (2) determine the behavior of fuels in a reactor system, and (3) study contamination problems by analysis of the activity distribution through the loop and decontamination devices, and by separation and characterization of measurable quantities of product. Two major advantages of this loop are that automatic in-line analyses of a variety of gaseous products and radioactive species will be possible with the instrumentation available, and a fully stainless-steel-lined gas-bearing compressor will be used to circulate gas in a chemically contamination-free environment.

Aerojet-General has operated a UO_2–liquid ammonia slurry in-pile loop but shut it down because of severe problems of slurry-fuel behavior and contamination. The Union Carbide Nuclear–Aerojet-General nuclear group is successfully operating a one-pass-flow fission-fragment fuel system at temperatures up to 600°C and a pressure of 34 atm.

Economic Evaluation

A number of capital- and production-cost estimates have been made on fissiochemonuclear reactors and processes. These are all subject to revision as new information and cost reductions are achieved in the civilian power-reactor programs in the United States and abroad. Firm estimates can never be made with a high degree of accuracy in a rapidly changing research field. About the only thing that can be done is to determine whether or not the results so far obtained are reasonable, and then to use these results for planning further research programs. Various indicators, or figures-of-merit comparisons, can be made (e.g., those in Table 2).

The competitive method is the synthesis of ammonia in an electrochemonuclear system. Hydrogen, produced in efficient low-cost water electrolyzers with d-c electrical energy generated by a nuclear reactor, can be combined with nitrogen, separated from air by liquefaction, in the conventional high-pressure catalytic process for ammonia synthesis. The equivalent G values and thermal-energy requirements per unit of fixed nitrogen calculated by using currently available efficiencies in low- and high-pressure electrolytic cells, combined with present average and maximum values for power-cycle efficiency, are given in Table 2. At a G value of 1 for the fission-fragment system, the energy requirement is very high. For an experimental G value of 6, the energy requirement is still 1.5 to 2.7 times as high as for the electrolytic system. Only at the kinetic maximum of 11.5 does the fission-fragment system begin to look competitive. Although there may be some advantage in plant cost for a direct fissiochemonuclear synthesis plant, the conclusion is that with the present state of knowledge it is difficult for the process to compete on the basis of producing nitrogen

Table 2 COMPARISON OF ENERGY REQUIREMENTS FOR FIXATION OF NITROGEN USING NUCLEAR ENERGY

Process	G value for N fixation	Energy-deposition efficiency, %	Net G value for N fixation	Energy required, kw(t)-hr/lb N
Fissiochemonuclear synthesis of NO_2 from atmospheric N_2 and O_2				
BNL experimental	1.0	50	0.5	174.0
Maximum experimental	6.0	50	3.0	29.0
Kinetic maximum	11.5	-	-	7.6
Thermodynamic maximum	284.0	-	-	0.31
Electrochemonuclear synthesis of NH_3 from electrolytic H_2 and atmospheric N_2				
Low-pressure cells (65% eff.)	14.7	30*	4.4	19.8
High-pressure cells (85% eff.)	19.2	42*	8.1	10.8
Dual-cycle synthesis, combining the electro process at 10.8 kw(t)-hr/lb of nitrogen with the fissio process at the indicated G value	1.0	50	0.5	10.2
	6.0	50	3.0	7.8
	11.5	-	-	4.5

*Thermal- to electric-power generation efficiency.

dioxide alone in a single-purpose plant. However, by combining a fissiochemonuclear system with power production in a dual-cycle system (Fig. 6), as shown in the bottom part of Table 2, a G value of 6 or a net G value of 3 might be economically attractive. The

cost factors into account, a generalized production-cost analyzer similar to one presented in Ref. 8 has been updated and expanded to include multipurpose chemonuclear systems.[32] The new formula for determining production costs reflects the considerably

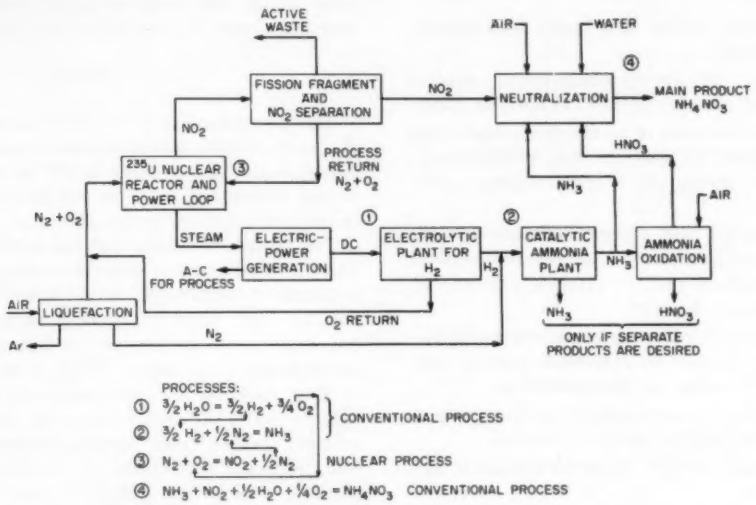

Fig. 6 Dual-cycle fissiochemonuclear and electrochemonuclear systems for nitrogen fixation.

total energy requirement could thus decrease to 7.8 kw(t)-hr per pound of fixed nitrogen and would then be about as low as the thermal equivalent of natural gas now being used to a large extent as the conventional source of energy and raw material for ammonia production. At a net G value of 3, 27% of the nitrogen would be fixed as nitrogen dioxide directly in the reactor at temperatures high enough to generate power for production of electrolytic hydrogen, which would be used in fixing 73% of the nitrogen as ammonia. It thus becomes necessary to determine the radiation yields at temperatures above about 500°C, where it is economical to convert thermal to electric power. The economics can be further improved in conjunction with large integrated multipurpose plants, which would produce fixed nitrogen and electrolytic oxygen in addition to desalinated water and power.

It should be noted that the concomitant development of low-cost high-current-density electrolyzers would materially aid in the economic development of the dual-cycle system.[31]

Energy requirements alone are not sufficient to determine the economics of a process. It is well known that nuclear power plants generally tend to have a lower fuel cost and a higher capital investment than comparable fossil-fuel plants. Taking all lower reactor costs found possible by the nuclear power industry within the last 2 years:

$$V = \frac{1216}{Ge_0M}\left[(0.0142fC_{DP} + 0.102) - F\left(1 - \frac{Ge_0}{G_{max.}}\right)(0.0142fC_E + 0.079) + (1 - f)\left(1 - \frac{Ge_0}{G_{max.}}\right)(0.0142fC_C)\right] - (1 - F)\left(1 - \frac{Ge_0}{G_{max.}}\right)\sum_{n=1}^{n}\left(\frac{G_{BE}e_P M_{BE}V_{BE}}{Ge_0M}\right)_E - \sum_{n=1}^{n}\left(\frac{G_{BF}M_{BF}V_{BF}}{GM}\right)_n \quad (6)$$

where V = production cost of primary fissiochemical product, cents/lb (excluding chemical raw-material cost)

G = G value of primary fissiochemical product, molecules/100 ev

e_0 = fission-energy-deposition efficiency (fraction of 200 Mev/fission deposited as radiation energy in the chemical system)

$G_{max.}$ = thermodynamic maximum G value for primary fissiochemical product, molecules/100 ev

f = fixed charge rate on capital investment (includes such items as depreciation, cost of capital, taxes, and insurance), a fraction

C_{DP} = unit capital cost of a dual-purpose (fissio and electric power) chemonuclear plant, \$/kw(t)

M = molecular weight of primary fissiochemical product

F = fraction of fissiochemonuclear reactor thermal power converted to electric power

C_E = unit capital cost of an electric power plant equivalent in size to the available by-product power for a chemonuclear plant, \$/kw(t)

C_C = unit capital cost of electrochemical (cells and rectifiers) and additional chemical equipment needed to produce electrolytic by-product, \$/kw(t)

G_{BE} = equivalent G value for electrochemical by-product (based on electrical energy consumed in cells), molecules/100 ev

e_p = efficiency of conversion of nuclear thermal power to electric power, a fraction

M_{BE} = molecular weight of electrochemical by-product

V_{BE} = electrochemical by-product credit cost, cents/lb

G_{BF} = G value for fissiochemical by-product, molecules/100 ev

M_{BF} = molecular weight of fissiochemical by-product

V_{BF} = fissiochemical by-product credit cost, cents/lb

n = nth by-product

The above general formula is based on the latest fuel and fuel-cycle cost quoted by the U. S. Atomic Energy Commission and reactor manufacturers. Capital-cost estimates are based on those now being quoted by nuclear-power-equipment manufacturers for light-water reactors,[33] and adjustments have been made for power-conversion equipment when estimating single-purpose, dual-purpose, and dual-cycle plants. In the estimates for electrochemical equipment, forthcoming improvements in cell construction and operation of water electrolyzers at high current densities[34] were assumed. A conservative assumption made was that the chemical recovery and decontamination plant investment is equal to the investment in the fissiochemonuclear reactor. The capital cost can be expressed as a function of the reactor power rating, P, in thermal megawatts:

$$C_{DP} = \frac{830}{P^{0.4}}\left(1 - 0.35\,\frac{Ge_0}{G_{max.}}\right) + \frac{500}{P^{0.4}} \qquad (7)$$

$$C_E = \frac{830}{\left[F\left(1 - \frac{Ge_0}{G_{max.}}\right)P\right]^{0.4}} \qquad (8)$$

When no power or electrochemical product is being produced, the former equation reduces to an expression giving the single-purpose fissiochemonuclear unit plant cost, C_S, in dollars per thermal kilowatt:

$$C_S = 1000/P^{0.4} \qquad (9)$$

The significant point to be emphasized here is that the unit capital investment has been markedly reduced in the past 2 years due to the use of new techniques in building reactors and the experience gained in operating nuclear power plants.

The above equations and fissiochemical G values already demonstrated experimentally or thought to be reasonably achievable were used in determining reactor and plant capacities at production costs currently possible for comparison with competitive conventional processes. Table 3 shows the results of this analysis for two types of processes: high-volume, low-risk utility-chemical systems and low-volume, higher risk industrial-chemical systems. For the highly competitive fixed-nitrogen and synthesis-gas cases, fixed charge rates comparable to public and private utility rates (7 and 14%) must be used to be reasonably competitive. This can be justified on the basis that fixed nitrogen for fertilizer and synthesis gas for pipeline distribution are fast becoming utility chemicals in the same sense as electric power and water are considered utilities today.

The fixed-nitrogen cases indicate that G(N) values in the range 6 to 11 in dual-cycle and dual-purpose plants are competitive with \$25 to \$30 per ton of equivalent ammonia, which is in the range of the lowest production cost possible today. The capacities required are in the range 500 to 5000 tons of fixed nitrogen per day. Conventional processes in this range of capacities are in existence today in single-plant locations, and thus the chemonuclear system is not unrealistic in this respect. A significant advantage of the chemonuclear plant is the long-term availability of fuel as plant sizes become larger. Consumption of fossil fuel in conventional plants becomes extraordinarily high, and feed stocks are rapidly depleted, with the result that costs increase. Larger chemonuclear plants can be shown to be more competitive; however, the equations for capital investment become less accurate. Credit for by-product oxygen under certain conditions is a significant factor. If electric power is more valuable than turning it into fertilizers, the dual-purpose case becomes more interesting than the multicycle cases. At higher G values there is less need for crediting by-products to hold costs down.

Table 3 CHEMONUCLEAR-PRODUCTION-COST ESTIMATES
(Raw Material for All Processes: Air and Water)

Product	System	G value for f.f. energy*	Energy,† kw(t)-hr	Fixed charge rate (f)	Reactor power (P), Mw(t)	Total plant cost, $ million	Plant capacity (T), tons/day	Production cost (V)
Fixed nitrogen in NH_3 + NO_2, O_2 by-product	Dual cycle	6	11.1/lb	0.07	2000	160	2150 N, 5750 O_2	\$30/equiv. ton NH_3, \$4/ton O_2
Fixed nitrogen in NH_3 + NO_2	Dual cycle	11	8.5/lb	0.14	3700	241	5250 N	\$30/equiv. ton NH_3
Fixed nitrogen in NO_2, electric-power by-product	Dual purpose	6	29.0/lb	0.07	1300	98	540 N, 400 Mw(e)	\$25/equiv. ton NH_3, 4.0 mills/ kw(e)-hr
Synthesis gas (CO + H_2), O_2 by-product	Dual cycle	20	2.9/lb [218/10^3 cu ft (STP)]	0.07	1380	105	5600‡ CO, [152 × 10^6 cu ft (STP) CO + H_2], 3290 O_2	\$0.30/10^3 cu ft (STP) H_2 + CO, \$4/ton O_2
Synthesis gas (CO + H_2), O_2 by-product	Dual cycle	10	4.0/lb [293/10^3 cu ft (STP)]	0.07	1880	133	5700‡ CO, [154 × 10^6 cu ft (STP) CO + H_2], 3290 O_2	\$0.38/10^3 cu ft (STP) H_2 + CO, \$4/ton O_2
Ozone	Single purpose	6	8.5/lb	0.14	423	38	600	\$47/ton
Hydrogen peroxide	Single purpose	1.7	42.0/lb	0.25	700	51	200	\$0.15/lb
Hydrazine	Single purpose	2.0	38.0/lb	0.25	100	16	30	\$0.25/lb (excludes NH_3 cost)

*f.f. = fission fragment.
†Energy requirement is total for product and by-products; includes fission-fragment and electric energy. Basis:

e_0 = f.f. energy-deposition efficiency = 0.50

e_P = thermal- to electric-power efficiency = 0.311

Energy for H_2 electrolytic cells = 140 kw(e)-hr/10^3 cu ft H_2 (STP)

‡Equivalent tons per day of CO; gas produced is CO plus H_2.

SYNTHESIS GAS

The CO- and H_2-gas-product cases have been computed only for the dual-cycle process, so that there is no credit for by-product power. All the power is internally consumed, producing H_2 and O_2 electrochemically. The results indicate utility costs of \$0.30 to \$0.60 per 10^3 cu ft (STP) for G(CO) values between 10 and 20. The plant capacities are limited to the capacity of a steel mill for iron-ore reduction because the steel mills can also use the by-product oxygen from the cells, which can be credited at the low cost of \$4 per ton of oxygen. These costs are in the range of being competitive with gas produced from coal. Hydrogen produced from natural gas costs about \$0.30 per 10^3 cu ft (STP) but is geographically limited. With the above equations it is possible to show that CO, H_2, and by-product O_2 can be produced in extremely large quantities for a minimum of \$0.15 per 10^3 cu ft (STP). These very large chemonuclear plants may be of use in providing gas supplies for distribution in the pipeline grids now in existence. The CO_2 feed to the chemonuclear plant can be supplied either by recovery from the consumer (i.e., iron-ore-reduction furnaces) or, if no other more concentrated source is available, from the atmosphere, which contains a vast store of CO_2 even though it is at a low concentration. As Table 3 indicates, heavy capital investments will be required to achieve the returns expected in these high-volume cases.

The other products shown in Table 3, which include ozone, hydrogen peroxide, and hydrazine, have the potential of being produced at significantly lower-than-present conventional production costs at plant capacities that are quite reasonable. The ozone case is for large-scale water treatment and pollution control.

The whole point of this economic study was not to obtain high accuracy but only to determine whether, under reasonably optimistic conditions, there is a chance for fissiochemonuclear processes to hold costs down in the future. As the supply of natural fossil fuels decreases, the cost of producing materials based on these fuels will certainly increase. The above analysis shows that, in the long run, even with present knowledge, chemonuclear processes are capable of producing large quantities of industrial and utility chemicals at costs that are not significantly higher than now possible.

OZONE

The possibility of producing ozone in chemonuclear reactors for water purification and pollution control

has some interesting implications. Ozone has been successfully used in the United States and France, among other places, for treating and purifying water without leaving unpleasant tastes and odors. Order-of-magnitude estimates made[35] for three different chemonuclear ozone plants for treating water for the New York City area are shown in Table 4. The Riverhead case is a small demonstration plant utilizing some of the reactor radiation escaping the swimming-pool core of the New York State dual-purpose power–water-desalting demonstration plant. Only enough ozone for treating the desalted water produced by the plant is necessary, and this can probably best be supplied by electrically powered conventional ozonizers. The ozone for treating the New York City drinking-water supply of 10^9 gal/day could be provided for by using a single-purpose (n,γ) chemonuclear plant. It would, however, make more economic sense if, in this case, the ozone was a by-product of an 1800-Mw(t) dual-purpose water-desalting–ozone plant producing approximately 180×10^6 gal of potable water per day. It should be noted that an (n,γ) reactor has the advantage of minimizing the contamination problem. When treatment of large bodies of water, such as the Hudson River, for purification and pollution control is considered, the hundreds-of-tons quantities of ozone required dictate the use of a fissiochemonuclear process, as shown for the last case in Table 4. Here the large amount of water treated, together with the increased concentration of ozone needed, rules out (n,γ) reactors, which would require hundreds of thousands of megawatts of thermal power to fill the demand. Hundreds of megawatts of fission-fragment power make the situation far more reasonable and economical than (n,γ) power or the use of multiplicity of low-capacity conventional electrically powered ozonizers. As indicated in Table 4, at a $G(O_3)$ value of 6 and 30 ppm O_3 for the Hudson River pollution requirement, it is estimated that a 600-Mw(t) reactor would treat 5×10^9 gal of water per day, producing ozone at an equivalent water-treatment cost of 7 mills per 1000 gal of water. Other situations, such as treating the Great Lakes and the Mississippi River water, can also be considered for this kind of application. The ozone case is a good illustration of the applicability of various combinations of chemonuclear systems depending primarily on capacity and multipurpose operation. It also serves to indicate the far-reaching scope of chemonuclear reactors when considering them as basic utilities rather than just as industrial-chemical producers.

Table 4 CHEMONUCLEAR REACTORS PRODUCING OZONE FOR WATER TREATMENT

[$G(O_3) = 6$]

Case	Water rate, gal/day	O_3 rate, lb/day	O_3 conc. in H_2O, ppm by wt.	Radiation	Efficiency, %	Power, Mw(t)
Riverhead	1×10^6	17	2	n,γ	0.5	2
New York City water supply	1×10^9	17×10^3	2	n,γ	0.5	1,800
Hudson River	5×10^9	12×10^5	30	n,γ	0.5	127,000
				f.f.*	35	600

*f.f. = fission fragment.

Contamination Problems

Although relatively little direct development work is being done on radioactive contamination of products, the problem is by no means being ignored. Capsule experiments have indicated residual contamination mainly from the halogens and noble gases. Product-distillation experiments containing radioactive tracers indicate that sufficiently high liquid decontamination factors can be achieved to yield products meeting acceptable health standards. Much process technology on the use of evaporation, filtration, distillation, and ion exchange for this purpose is available from operation of existing nuclear-fuel-processing plants. None of the problems appear to be technically insurmountable. However, before a large-scale development effort on decontamination is justified, it is first necessary to demonstrate the attainment of economic yields with a fair degree of certainty. Following this, sufficient pilot quantities of product should be produced to characterize the contamination problem. Upon establishing the extent of the problem, a meaningful solution can then be developed.

Consideration of the above experimental and projected results in the light of the nation's and world's population growth, together with the food, water, and fuel supply, leads to the conclusion that there is more than ample incentive—if not for private industry, then for the nation—to support a continued orderly investigation of both the direct and indirect conversion of nuclear to chemical energy.

Radiation Chemical Processing

Let us turn now to the use of isotopic and machine radiation sources for inducing exothermic, chain, reactions for chemical synthesis. Those that have been

more prominently mentioned in the literature are listed in Table 5. The reasons and incentives for considering radiation induction vs. other methods are: (1) the reaction can be carried out more conveniently with radiation; (2) the product cannot be easily produced by other methods; and (3) the process is more economical. The requirement that chain reactions must be exothermic establishes the type that must be considered, i.e., halogenation, polymerization, partial oxidation, and catalytic reactions. The competitive processes include thermal, thermocatalytic, and photolytic induction.

The only known radiation chemical process that has reached commercial status is the relatively small-capacity Dow process for hydrobromination reported as being produced by thermocatalytic methods. The formation of wood–plastic combinations by in situ radiation-induced polymerization of liquid monomer with which wood has been impregnated has caught the fancy of many domestic and foreign organizations.[39]

In the category of oxidation reactions, the addition of metallic ions to an aqueous benzene solution has recently been shown to increase the radiation yield markedly at temperatures lower than those attainable by purely thermal induction.

The possibility of improving and extending the life of catalysts, for example, in ammonia synthesis and in electrode processes by irradiation, remains an open area for research.

Table 5 RADIATION CHEMICAL REACTIONS

Reaction*	Product	Heat of reaction at 25°C, kcal/mole	G_{exp}
Halogenation			
$C_2H_4(g) + HBr(l) = C_2H_5Br(l)$	Ethyl bromide	−24.2	39,000
$C_6H_6(l) + 3Cl_2(g) = C_6H_6Cl_6(l)$	Hexachlorobenzene		85,000
Polymerization†			
$n(C_2H_4)\ (g) = (C_2H_4)_n(s)$	Polyethylene	−22.0	10^5 to 10^6
$n(C_6H_5-C_2H_3)\ (l) = (C_6H_5-C_2H_3)_n(s)$	Polystyrene		10^6
$n(CH_2=O)\ (s) = (CH_2=O)_n(s)$	Polyester		
Partial oxidation			
$2RH(l) + 2SO_2(g) + O_2(g) = 2RSO_2H(l)$	Alkane sulfonate		5,000
$C_6H_6(l) + \frac{1}{2}O_2(g)\ (Fe^{2+}) = C_6H_5OH(l)$	Phenol	−1.9	1,000

*Here (l), (g), and (s) refer to liquid, gaseous, and solid states.
†G_{exp} values for polymerization reactions are in terms of number of monomer molecules converted to polymer per 100 ev absorbed in monomer.

of ethylene. It is understood that chlorination of benzene for hexachlorobenzene production was considered for commercialization but that the market was insufficient to support production from a new plant. It has also been reported that the sulfoxidation of long-chain hydrocarbons is near commercialization in connection with the production of biodegradable sodium alkane sulfonate detergents.

Radiation-induced polymerization of monomers has been under study for a number of years. Some of the latest results on gas-phase polymerization of ethylene, which has been pursued by many investigators, are presented below. The only new departure, known to the writer, on liquid-phase polymerization is the recently discovered extremely high radiation-induced polymerization rate of extremely dry styrene.[36] A solid-state radiation-polymerization process for making polyester from trioxane has reportedly been developed in Japan. Emulsion polymerization of vinyl acetate to produce high-molecular-weight material has some interesting features.[37] Along the lines of unusual processes and products, the copolymerization of ethylene with low-cost raw materials such as carbon monoxide and sulfur dioxide can be mentioned.[38] These are high-molecular-weight polymers with good thermal stability which have not yet been

Probably the main reasons for lack of rapid application of high-energy radiation to chain reactions include: (1) a lack of radiation processing know-how in the industry; (2) the more often than not marginal economic advantage of a radiation process over other methods; and (3) the slow and expensive rate of introduction of a new radiation-synthesized product.

The radiation-induced homopolymerization of ethylene has all the elements of an ideal process for demonstrating and developing the technology of a large-scale radiation chemical process. Capsule research has indicated that the mechanism is, in the main, free-radical controlled. The polymerization is inhibited by oxygen and is dependent on approximately the half-power of the radiation intensity. Below the polymer melting point (20 to about 120°C), the rate is practically independent of temperature; however, above the melting point (120 to 200°C), the activation energy is approximately 14 kcal/mole and the rate increases rapidly. The rate of polymerization is strongly dependent on pressure, and at higher temperatures and above 850 atm the pressure may have a higher exponent than indicated in Table 6 (> 2.7). Under the latter conditions, rates greater than 1 g/liter · hr/atm have been obtained, which is in the

Table 6 COBALT-60 GAMMA-RADIATION-INDUCED POLYMERIZATION OF ETHYLENE
(Results at Limits of Experimental Conditions in Static Capsule Experiments)

Intensity (I), rads/hr	Pressure (P), atm	Av. temp., °C	Conversion, %	Rate (Q),* g/(liter)(hr)	G value,* molecules/100 ev
182,000	1920	21	8.0	593	189,000
847,000	680	32	70.5	780	64,000
179,000	850	200	13.1	2,870†	1,130,000

*Run terminated by decomposition reaction. The equations for 20 to 100°C, 170 to 1360 atm, and (0.58 to 8.5) × 10^5 rads/hr are:

$$Q = 1.39 \times 10^{-6}\, e^{-200/RT}\, p^{1.7}\, I^{0.5} \pm 27\%$$

$$G = 3.5\, e^{-200/RT}\, p^{1.5}\, I^{-0.5} \pm 27\%$$

For the temperature range 120 to 200°C, the activation energy E = 14,500 kcal/mole.

range of conventional rates. Conditions are such that explosive decompositions have been encountered.

Analysis of the product, Gammathene, indicates that the material produced above the melting point is a lower density high-melt-index soluble polymer, similar to the film-grade polyethylene produced by conventional high-pressure processes. Gammathene produced below the melting point is a high-density high-molecular-weight long-chain linear polymer with a low melt index. This material may be considered by some as a new product in the polyethylene market.

A preliminary design and evaluation study has been made on the radiation conversion of ethylene,[40] assuming that the gas-preparation and polymer-finishing parts of the process remain similar to those in the conventional system. The reaction vessel is a high-pressure cylinder with a central source. For a constant-capacity plant, there is an optimum, which is controlled by, among other factors, the cobalt and vessel costs. This optimum occurs because, although the energy loss increases with increasing intensity, the conversion rate also increases, and a smaller, and thus cheaper, reaction vessel can be used. The optimum for a high-temperature plant with a capacity of 5×10^7 lb/year is at 680 to 1360 atm pressure for a temperature of 200°C and permits a significant decrease in investment and production costs (~0.6 cent per pound) over conventionally manufactured low-density polymer (Fig. 7).

The low-temperature Gammathene may also be capable of being produced at costs comparable to conventional ones. A 100-ml-volume experiment with flowing gas is being operated now at BNL to confirm the rate equations and to produce sufficient polymer for commercial evaluation by industry. These results indicate that there is ample incentive for developing a radiation process which the nation can make use of in the greatly expanding polyethylene market and which can also serve as a demonstration for gaining experience when considering other more individually based industrial processes. (MG)

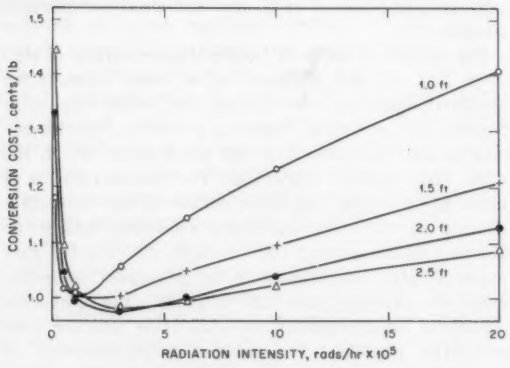

Fig. 7 Conversion cost as a function of radiation intensity and vessel diameter in 5 × 10^7 lb/year plant for radiation-induced polymerization of ethylene. Operating conditions: 680 atm, 200°C. Return rate, 15%; conventional conversion cost, 1.56 cents per pound.

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Section

V

Isotopes and Radiation Technology

Source Development

Large Scale Production and Applications of Radioisotopes*

Abstract: The proceedings of the American Nuclear Society National Topical Meeting on large-scale production and applications of radioisotopes, held at Augusta, Ga., Mar. 21–23, 1966, are summarized. In addition to three introductory addresses, papers were presented in sessions on large-scale (1) production of radioisotopes, (2) chemical processing, (3) international programs, (4) applications of radiation, and (5) applications of isotopic power.

Information presented at the meeting* on large-scale production and applications of radioisotopes is reviewed. The speakers, whose papers appear in Vol. I, were invited to discuss subjects in which they are acknowledged experts. Papers covering the work of other scientists on any aspects of the subject of the meeting had been requested previously on an international basis. All submitted papers, published in Vol. II, were summarized by the invited speakers, who acted as rapporteurs. About 400 people attended the meeting.

This review is arranged under the same subject categories as those of the Proceedings, i.e., addresses and sessions on large-scale (1) production of radioisotopes, (2) chemical processing, (3) international programs, (4) applications of radiation, and (5) applications of isotopic power.

Introductory Addresses

Opening remarks by Dessauer,[1] E. I. du Pont de Nemours & Co., Inc., stressed that isotope production and use is very likely to develop into a large industry by 1980, if not sooner. For example, the predicted power reactors will have the capacity to produce ^{237}Np and ^{243}Am in sufficient quantities to make the recovery of these materials worthwhile for further conversion to ^{238}Pu and ^{244}Cm in reactors optimized for neutron economy. At the same time, these latter products, and many others, will be needed in at least the predicted quantities for applications in the oceans, on land, and in space—to all of which they bring unique advantages over alternative sources of power or radiation. Therefore efficient production methods must be developed, and enough of the candidate materials must be made available in existing production facilities to prove and perfect the devices in which the isotopes are to be used.

Fowler,[2] Division of Isotopes Development (DID), AEC (paper presented by J. E. Machurek), reported that, although the total market of the process radiation industry has reached approximately $125 million per year, most of us believe that the few processes now in operation are merely forerunners of what is to come. Studies indicate that this industrial potential is significant. Possible explanations of why a more rapid growth pattern has not been demonstrated are the lack of trained personnel in industry; a reluctance to exchange conventional processes for radiation applications based on insufficient operating experience; some early failures combined with overstatements; the early high cost of sources; the continual changes in radiation cost which make investment speculative; and a general public fear of radiation and atomic energy.

Today not enough proven commercial radiation processes exist to support an intermediate-scale process radiation industry that can pull together the necessary talents and capabilities to serve industrial needs. To overcome these obstacles and accelerate the growth of a real process radiation industry, the DID has designed its Process Radiation Development Program to accomplish missions in three areas:

1. Basic Radiation Technology: to broaden basic technology, particularly in radiation chemistry. The long-range program will concentrate on strategic areas of research so that new commercial processes can be built on a firm basis of understanding.

2. Radiation Process Engineering: to develop the basic "hardware" for process radiation and obtain engineering process data. Much of this information is being compiled and will be distributed in a Radiation Engineering Handbook.

*Large Scale Production and Applications of Radioisotopes, Proceedings of American Nuclear Society National Topical Meeting, Vols. I and II, March 21–23, 1966, Augusta, Ga., USAEC Report DP-1066 (Vols. I and II) (CONF-660305), E. I. du Pont de Nemours & Co., Inc., Savannah River Laboratory, May 1966. (Available from the Clearinghouse for Federal Scientific and Technical Information, National Bureau of Standards, U. S. Department of Commerce, Springfield, Va. Price, $3.00 each volume.)

3. Applied Cooperative Industry/AEC Projects: to provide private industry an incentive for investment by means of demonstration projects. This program, it is hoped, will encourage investment dollars in process radiation and permit the AEC to withdraw its development effort after industry has indicated it is capable and willing to take over the process. Three cooperative demonstration projects under consideration are meat irradiation, production of wood–plastic materials using radiation to polymerize a monomer that has been absorbed in wood, and production of polyethylene and related copolymers.

The Department of the Army has been conducting research on the sterilization of meat products, and this work has progressed to a development-program stage. Private facilities do not exist for producing the large quantities of radiation-sterilized meat required. However, the AEC is interested in fostering both the commercialization of radiation-preserved foods and the design and construction of radiation facilities, and both the AEC and the Department of Defense (DOD) would prefer maximum private participation. Since the Army will require large quantities of radiation-sterilized meats for its development program, DOD and AEC have proposed the following plan:

1. Industry will be invited to submit proposals for the design, construction, and operation on a commercial basis of the proposed meat irradiator.
2. The DOD will commit itself to purchase from the operator specific quantities of irradiated meats annually.
3. The AEC will provide, without cost, a radiation source and design assistance.

With these cooperative programs in which industry and government share the cost and risks, the AEC hopes to (1) demonstrate the practicability of using isotopes and radiation technology, (2) further develop the peaceful uses of atomic energy, (3) reduce AEC development costs, (4) reduce economic risks to a level more in line with those of conventional processes, (5) provide markets for radioisotopes and electron accelerators, and (6) enhance the national economy and welfare.

Spinrad[3] of Argonne National Laboratory (ANL) gave long-range speculations on isotopic power at a luncheon address. He recalled that radioisotope power had been talked about for more than 60 years and then questioned what the ultimate future holds. To understand this we must first try to think what radioisotope sources can do that other sources cannot.

The outstanding characteristics of an isotope source are that it is completely self-contained, it does not change except for decay, and it does not require control to get a predictable output. On the negative side, the power cannot be shut off, it diminishes with time, and radiation from the source sometimes presents shielding problems. In competing with chemical fuels, radioisotopes have the advantage for any mission of more than 500 hr, and eventually they will probably be the choice for any mission over 100 hr. With nuclear reactors, isotopes compare favorably only where the reactor fuel is used just once and cannot be reprocessed — for example, in space. However, there is a limit to the minimum size of a reactor; below 20 to 100 kw, improved isotope sources will probably be preferable.

Beyond the immediate future, isotopes whose manufacture consumes reactor neutrons do not seem competitive with by-product isotopes, which, wanted or unwanted, are normally produced during reactor operation and are the cheap nuclides. Americium-241 may be considered a natural by-product, but ^{238}Pu is not because it is produced by neutron irradiation of its chemically separated precursor, ^{237}Np. The chief by-products are the fission products; however, others — ^{242}Cm and ^{243}Am — will come from long-burning reactor fuels. On the other hand, ^{244}Cm must be considered as "manufactured." In a world where nuclear power is becoming quite prevalent, the reactor by-product radioisotopes will be very abundant and relatively cheap, but the manufactured isotopes will cost as much as new fissile nuclides: $20 thousand to $30 thousand or more per mole.

What, then, are the future applications for these isotopes? Sites where small isotopic power sources are now used are defined by today's standards as relatively inaccessible parts of the earth (e.g., buoys and arctic stations), but these will probably not be so inaccessible in the future and replacement will be less difficult. Where, then, will the demand be? The entire ocean floor (the "benthic" region) remains a very important possible application for isotopic power for navigational markers, with space needs the second largest demand.

By-product isotopes may be matched to missions by half-lives in essentially three groups:

Half-life, years	Nuclide
1	^{144}Ce
2	^{147}Pm, ^{125}Sb, ^{155}Eu
30	^{90}Sr, ^{137}Cs

To fill the ranges of half-lives not covered by these nuclides, we have to look at the manufactured isotopes or marginal by-product alpha emitters:

Half-life, years	Nuclide
0.5	^{170}Tm, ^{242}Cm
5	^{60}Co
18	^{244}Cm
80	^{238}Pu
460	^{241}Am
1620	^{226}Ra

Spinrad then matched missions to the half-life of a particular nuclide used in a power source. Radium-226 and ^{241}Am would be used for essentially "permanent" systems, e.g., for the outer reaches of the solar system or for repeater relays for deep-space probes. Shorter lived power sources with half-lives of about 30 years could be used for systems involving multiprobes fired one per year to map magnetic forces, plasma density, or any other form of radiation in the solar system. These probes would report periodically and simultaneously over a long time. For shorter space missions, ^{60}Co would be about right for the 5-year round trip to Jupiter; ^{147}Pm, for the 1-year round trip to Mars; and ^{242}Cm, for the half-year Poodle outbound trip to Mars.

In the not-too-distant future, sonic navigational markers will be required for oceanographic and submarine shipping lanes, and, to generate a decent sonic signal, a large amount of power is required. Cobalt-60, ^{244}Cm, or, if replacement becomes feasible, a 2-year fission product may be used.

Spinrad concluded that, although isotopic power costs and economics are important, in the impact of isotopes on society, isotopic power must take second place to the myriad of uses of isotopes for tagging and investigating. Nothing else has been able to do the job that isotopes do here, and no matter how small the volume of research and tagging business, these uses are the most important ones for us.

Tape,[4] an AEC Commissioner, discussed basic research and development in the search, identification, and production of the transuranium elements and the utilization of reactors for direct production of isotopes. The year 1966 is a most propitious time to consider the development of ^{238}Pu, a material for which there will be many large-scale applications. Twenty-five years ago this isotope was first identified as a product of bombardment in the 60-in. University of California cyclotron. A few weeks later, fission of another isotope of this element was observed when it was bombarded with neutrons. Today 11, or possibly 12, transuranium elements are known.

Nuclear power plants built, being built, or on order in the United States will produce more than 3 thousand kg of plutonium per year, and, with the projected growth of nuclear power, the potential U. S. annual production should be about 20 to 30 thousand kg per year by 1980. These same plants will produce ^{237}Np, the precursor of ^{238}Pu, which is currently favored for use in the space program. Power reactors plus AEC resources could produce about 15 hundred kg of ^{238}Pu by 1980.

The Commission is pursuing many avenues for production and study of transuranium elements: intense neutron irradiation in very-high-flux reactors, heavy-ion bombardment in special-purpose accelerators, and instantaneous neutron irradiation in nuclear explosions. A National Transuranium Program was established in 1957 with the irradiation of 20 kg of ^{239}Pu in a Savannah River production reactor to produce ^{242}Pu, ^{243}Am, and ^{244}Cm, the essential materials for further production of heavier elements. The second step was the construction of the High Flux Isotope Reactor (HFIR) and a Transuranium Processing Plant (TRU) at Oak Ridge National Laboratory (ORNL).

The Commission is also interested in the beneficial use of fission products and is pleased that industry has enough confidence in the field of isotopic power and radiation applications to invest over $8 million in the processing facility at Hanford. This plant should provide tens of megacuries of ^{90}Sr, ^{137}Cs, ^{144}Ce,* and ^{147}Pm.

In considering reactor products the AEC feels that, over the long term, industry will have a major role in the production of such isotopes as ^{238}Pu and ^{244}Cm. With the large quantities of isotopes that will be produced in power reactors, it is important that this isotopic energy not be wasted but be put to use and that a market for these products be developed. The AEC can play a significant role not only in its development program but also by producing useful quantities for first-generation applications.

The AEC has increased its knowledge of the capability of the production-reactor system to produce various isotopes while still fulfilling the weapons mission. As an example, ^{60}Co with a specific activity of 700 curies/g has been made at Savannah River, and one of the production reactors, in a demonstration of high-flux operation, reached a peak flux level of 6×10^{15} n/(cm^2)(sec) in a volume of about 165 cu ft.

Polonium-210 is receiving considerable attention, and indications from the National Aeronautics and Space Administration (NASA) are that future needs for ^{238}Pu may be great enough to require production in commercial power reactors as well as in production reactors.

Large-Scale Production of Radioisotopes

IN PRODUCTION REACTORS

Many of the coauthors with Overbeck[5] — at Hanford and Savannah River, in Wilmington, and in Washington — requested that he express their views forcefully. Four points summarize these views:

1. The need for radioisotopes is urgent and substantial.

2. We must learn to interpret these needs realistically and determine what they mean in terms of production facilities and production schedules.

*Recent information indicates that ^{241}Am will be produced in place of ^{144}Ce; however, the ^{144}Ce process will be included in the license agreement in the event that a need for this nuclide develops at a later date.

3. Efficient large-scale production of isotopes requires a substantial effort in process development. Work is under way, but much remains to be done. Paper studies alone are not adequate.

4. Work is being done on methods to produce all the interesting isotopes, but the most urgent needs may have to be filled with the isotopes that are easiest to produce.

Missiles and Rockets[5a] reports that NASA's needs for ^{238}Pu may reach the equivalent of 250 kw(t) per year by 1980. Overbeck translates this need to more than a thousand megawatts of production capacity. If other isotopes are also needed, several thousand megawatts of production capacity may be required. The ability to meet such demands exists only in the AEC-owned production reactors at Hanford and Savannah River.

To efficiently produce isotopes other than ^{239}Pu in the production reactors would require a shift toward enriched ^{235}U fuel and replacement of all or part of the ^{238}U target with some other target material. Such a diversion of capacity may range from incidental production, where a small region of the reactor might be altered to accommodate special target materials, to a full-scale production, where all the ^{238}U would be replaced by another target material. Overbeck then described the capabilities of the reactors at Savannah River and Hanford for isotope production.

A primary objective of the current pilot programs is to overcome the need for long lead times. Development work and experience gained now will save a great deal of time and money when urgent, full-scale demands arise.

Data in Glasstone's *Sourcebook*[5b] indicate that with a 1-thousand-Mw reactor there might be a maximum of 2 kg of productive neutrons from fission of about 1.3 kg-atoms of ^{235}U per year; these neutrons will produce 2 kg-atoms of any isotope that requires only a single neutron capture. Alpha, beta, or gamma emitters may be produced, their stored energy being directly proportional to the disintegration energy. Examples of power from various emitters are:

Produced by 2 kg of neutrons per year		
^{238}Pu	5.5 Mev α,	300×10^6 kw(t)-hr
^{170}Tm	0.3 Mev β,	16×10^6 kw(t)-hr
^{60}Co	2.6 Mev γ,	140×10^6 kw(t)-hr
Produced by fission of 1.3 kg-atoms of ^{235}U per year		
^{90}Sr	6.0% yield	1.150 Mev β, 2.400×10^6 kw(t)-hr
^{147}Pm	1.5%* yield	0.067 Mev β, 0.035×10^6 kw(t)-hr

The *Sourcebook* calculations, although not so refined as desired, indicate that (1) the most important function of a production reactor is to provide neutrons (in contrast, the power-reactor operator must sacrifice neutrons to efficiently produce directly useful power); (2) alpha emitters are favored because they give the most stored energy per neutron consumed, although ^{60}Co is a close second choice and is readily available; and (3) the fission products turn out to be less important than the neutron-produced isotopes from a production reactor since the anticipated needs for isotopic energy cannot now be met with fission products alone.

Overbeck concluded with a discussion of four source materials — ^{60}Co, ^{238}Pu, ^{244}Cm, and ^{210}Po. He included estimates of price per thermal kilowatt-hour, production capabilities and difficulties, and activity and required shielding. Although not a current candidate as an energy source, ^{252}Cf was also discussed briefly.

*Reduced from 3 to 1.5% to allow for decay of unwanted ^{148}Pm.

IN POWER REACTORS

After listing criterions that radioisotopes should meet for production in a power reactor, Rupp[6] stated that power reactors seem unlikely to be either needed or well-adapted for producing a wide range of radioisotopes for use in science and medicine. One should be careful, however, to exclude possible military or national defense uses, which were outside the scope of the meeting.

By-products, to use the term in the usual sense with nuclear reactors, would be only the fission products and certain transuranium isotopes that occur unavoidably during operation of the reactor for its main purpose. In optimizing parameters for production of one product or another, including power, one does not think of utilizing a by-product, but rather of designing equipment and operations for multiproduct production to yield the maximum amount of return on investment.

The immediate objectives of producers of commercial radioactive products and of producers of power (except for maximum dollar return on investment) are quite different. The power producer would be quite happy if he produced high-temperature heat energy and nothing else; the special isotope producer considers the heat a nuisance, which costs money to get rid of. Reconciliation of these different viewpoints and objectives is of main concern in using power reactors for radioisotope production; the connecting link between the two is the possibility of increasing return on investment.

The papers[6a–6e] reviewed by Rupp concerning use of power reactors for radioisotope production considered (1) ^{238}Pu and its precursors, primarily ^{237}Np; (2) ^{244}Cm and, to some extent, ^{242}Cm with plutonium and americium as precursors; and (3) ^{147}Pm and ^{210}Po. The production of ^{237}Np, ^{238}Pu, and, to a lesser extent, ^{244}Cm are described in some detail with emphasis on economies of fuel recycling, use of high-flux converter reactors, and special targets and reactor loadings to take advantage of

more favorable neutron spectrums. He pointed out that small amounts of ^{236}Pu contaminate ^{238}Pu, and, since this nuclide and its daughters emit gamma rays, it could disturb the preeminent position of ^{238}Pu as an isotopic fuel for gamma-free environments.

Rupp also mentioned that large-scale production of low-specific-activity ^{60}Co in production reactors should not be overlooked. Rupp's brief summary was:

1. Projected demand for all isotopic power fuels appears to go beyond the anticipated production (especially for ^{238}Pu).
2. The commercial nuclear industry can contribute substantially to supplying these isotopes, power reactors being complementary to production (converter) reactors.
3. Careful recycling of power-reactor fuels and integrated management of ^{236}U, ^{237}Np, ^{238}Pu, ^{241}Pu, $^{241-243}Am$, and ^{244}Cm will be necessary to optimize production of power fuels and ensure profits.
4. Commercial chemical processing and target fabrication of ^{237}Np and the actinides will be needed eventually.
5. Power reactors are unlikely to be good converter reactors; multipurpose multiproduct converter reactors of optimized design should be envisioned for the future.

IN COMMERCIAL REACTORS

Slosek[7] of the General Electric Company reported that prior to 1959 there was no domestic commercial production of radioisotopes since there were no suitable privately owned reactors. Private production of radioisotopes was not initiated until the General Electric Test Reactor (GETR) came on line. Today commercial producers fall into two large categories: those who operate their own nuclear reactors and those who lease space in commercial reactors for radioisotope production. In these two categories there are now 15 to 20 private firms who routinely produce and distribute over 30 nuclides.

Of the isotopes formed by neutron capture, ^{192}Ir, ^{60}Co, and ^{170}Tm have been produced commercially on a large scale. Iridium-192 is used predominately in radiography. It was first produced commercially in 1960, and its production currently exceeds 100 thousand curies/year. Specific activities of 200–250 curies/g are adequate for usual radiographic work, but material up to 400 curies/g has been produced commercially.

Cobalt-60 used for radiography is estimated at less than 50 thousand curies/year, radiotherapy requirements are less than 500 thousand curies/ year, and process radiation markets are difficult to define, since even a single new installation can have a significant impact on total requirement. The demand for ^{60}Co isotopic power sources is still uncertain, and AEC is conducting programs investigating such a use; if the results of the studies are favorable, potential requirements could be large and specific activities high.

The U. S. consumption of ^{60}Co during 1964 and 1965 increased substantially; the major portion being on new process radiation facilities. The AEC supplies low-specific-activity cobalt in large quantities for this application. The commercial ^{60}Co producer is closely following the emergence of the new market for process radiation.

Slosek then examined the economic feasibility of a reactor whose primary function is the production of ^{60}Co. Medium- and high-specific-activity ^{60}Co in sufficient quantities to meet current and projected requirements for radiotherapy and radiography applications is available from private sources on a routine basis. With AEC encouragement these applications have led to an industry that includes ^{60}Co producers, source encapsulators, and equipment manufacturers. If the process radiation market grows significantly and ^{60}Co can compete successfully with fission products in the market, additional private participation through production in some power reactors and commercial isotope-production reactors can be expected.

Thulium-170 was the first isotope to be produced in large quantities in commercial reactors. Approximately 150 thousand curies of ^{170}Tm was produced in 1959 and used to power an experimental thermionic converter. This work, done at General Electric's Vallecitos Nuclear Center, was one of the first direct energy-conversion experiments that used a radioisotope heat source; this was also one of the first times that more than 200 watts of thermal isotopic power was packaged in a single sealed source for any purpose. This isotope is particularly suited to prototype experiments: it can be prefabricated while nonradioactive and produced in small or large quantities on short notice; it is inexpensive; and it possesses characteristics of interest to developers of direct energy-conversion systems. Multikilowatt quantities of ^{170}Tm in fuel forms capable of delivering up to 10 watts(t)/cm^3 could be produced in existing commercial facilities on relatively short notice.

There has been no significant production of alpha-emitting isotopes in private reactors. Industrial capabilities exist in this area, however, and industry will use its facilities and technology to assist the government in any capacity requested.

SESSION DISCUSSION

In the discussion that followed the session, the utility of ^{60}Co as a power source was questioned because of its high energy and the shielding required. In answering this question one must ask: With what is ^{60}Co being compared? For large power generators

the weight of a ^{60}Co system could very well be less than that of a ^{90}Sr system shielded to the same tolerance level. High specific activity of a ^{60}Co power source would also considerably reduce the weight of shielding. Strontium-90, a by-product, will be available for high-energy heat sources with low-weight shielding without a great deal of further research and development, and it is intended primarily for terrestrial use. Cobalt-60 power systems were briefly compared with the SNAP reactor systems. Other radioisotopes cannot be compared with SNAP reactors on a realistic basis at this time because sufficiently large quantities of other radioisotopes do not exist. Discussions of ^{170}Tm included cost, purity, and supply of target material and the production, cost, and radiochemical purity of the product ^{170}Tm.

Large-Scale Chemical Processing

NEUTRON-PRODUCED ISOTOPES

Lamb,[8] reporting on the large-scale processing and source preparation of neutron-produced isotopes, i.e., those produced by the (n,γ) reaction, defined the term "large-scale processing" to be quantities in excess of 10 thousand curies/year. Table 1 indicates the potential demand for these isotopes as compared with the demand during the calendar year 1965.

Table 1 LARGE-SCALE PROCESSING OF NEUTRON-PRODUCED RADIOISOTOPES FOR U. S. COMMERCIAL AND ISOTOPIC POWER APPLICATIONS

Radioisotope	1965 usage, curies/year	Potential demand, curies/year
^{60}Co	1×10^6	$>1 \times 10^8$
^{3}H	3×10^5	$>3 \times 10^5$
^{238}Pu	2×10^5	$>4 \times 10^5$
^{192}Ir	1×10^5	$>1 \times 10^5$
^{242}Cm	4×10^4	$>3 \times 10^5$
^{244}Cm	1×10^4	$>3 \times 10^5$
^{210}Po	1×10^4	$>1 \times 10^8$
^{171}Tm	0	$>1 \times 10^6$

Five papers[8a–8e] dealing with ^{242}Cm, ^{244}Cm, and ^{171}Tm were used by Lamb to illustrate the operations involved in processing and in preparing sources of neutron-produced isotopes. All the isotopes in Table 1 except ^{60}Co and ^{192}Ir require rather complicated chemical processing and source-forming operations.

Purity of target materials is very important because unwanted activities may result from neutron reactions with the impurities. When the elemental form cannot be used as the target, the target compound selected should contain a high percentage of the target element, be stable under the heat of radiation, and contain only combined elements that yield no undesirable activities. Also to be considered is the selection of the container and matrix materials for easy postirradiation processing, heat generation in the target during irradiation, and void volume in the capsule to accommodate helium and fission gases produced.

Neutron-produced isotopes are recovered and purified using conventional solvent extraction, ion exchange, distillation, and precipitation processes. Unconventional effects, e.g., of radiolysis which causes acid depletion, change in valence of metal ions and anions, and reagent damage, are important considerations in large-scale radiochemical processing. Reagents are a potential source of impurities, and good quality control must be exercised to prevent introducing impurities that may reduce activity concentrations or diminish product quality.

The product must be in a highly stable solid form. This requires conversion to a compound or to the element, mechanical forming, and encapsulation to prevent release of contamination under predicted normal or abnormal operating conditions.

Lamb presented in some detail the ^{242}Cm and ^{244}Cm processing experience at ORNL[8c] and development of processes for ^{244}Cm and ^{171}Tm at the Savannah River Laboratory.[8b,8d] At the time of this meeting, 35 g each of ^{244}Cm and ^{243}Am and 12 g of ^{242}Cm had been recovered at ORNL. The objectives of this program are to test equipment, develop chemical processing flow sheets, and produce (1) pure ^{244}Cm for basic nuclear research and for use as reactor target material for the production of higher atomic number transuranium elements and as material for isotopic power development and (2) ^{242}Cm for isotopic power development work. In the future we can expect the processing scale for neutron-produced isotopes to be increased by factors of tens or hundreds, and we can anticipate refinement of the four basic process operations to permit economic production of these isotopes on a very large scale.

The discussion that followed Lamb's paper centered first on the isotopic purity of product ^{171}Tm and the target material ^{170}Er and then on the increased demand for ^{210}Po.

SEPARATED FISSION PRODUCTS

The processing of separated fission products and preparation of sources from them were described by Cochran.[9] Fission-product recovery was initiated at Hanford in August 1960 with the production of ^{90}Sr and ^{137}Cs. The program has since included production of custom quantities of ^{144}Ce and ^{147}Pm as well. The current program involves separation of strontium and rare earths from Purex-process acid waste by sulfate precipitation with a lead carrier and subsequent separation of rare earths from strontium by oxalate precipitation. Strontium is further purified by solvent extraction with di(2-ethylhexyl) phosphoric acid. Cesium-137 is recovered from aged alkaline

wastes by adsorption from the alkaline supernatant solution on a bed of aluminosilicate resin. Other processes that have been demonstrated in plant equipment are (1) ^{137}Cs recovery from alkaline wastes by ion exchange on zeolite followed by nickel ferrocyanide precipitation and from acid wastes by phosphotungstic acid precipitation; (2) ^{99}Tc recovery and purification by anion exchange; (3) ^{144}Ce separation from the rare earths by di(2-ethylhexyl) phosphoric acid solvent extraction using silver-catalyzed persulfate oxidation of Ce^{3+} to Ce^{4+}; and (4) subsequent ^{147}Pm separation from the cerium-free rare earths by chromatographic ion exchange.

A Fission Products Conversion and Encapsulation plant is being designed by Burns and Roe, Inc., that will have an estimated annual production capacity in excess of 25 Mc of each of the isotopes listed above except ^{99}Tc. The conversion processes, anticipated packaged forms, and processing steps for each isotope from receipt of the feed stream to shipment of the final isotope product were outlined at the conference. Production of sources in megacurie quantities is hoped for by 1968.

CHEMONUCLEAR AND RADIATION PROCESSES

The paper on chemonuclear and radiation chemical process research and development by Steinberg[10] is presented in its entirety beginning on page 142 in this issue.

Large-Scale International Programs

Five of the papers presented in this session will be reviewed in the next issue of this Technical Progress Review in the section "Isotopes Around the World." Programs reviewed will be those of Canada,[11] United Kingdom,[12] Continental Europe,[13] Japan,[14] and the International Atomic Energy Agency.[15]

The production and application of ^{227}Ac for direct-conversion energy sources, which is viewed as a specific Belgian contribution to the European space program, was described by De Troyer.[16] Actinium-227 is produced by reactor irradiation of ^{226}Ra by the $^{226}Ra(n,\gamma)^{227}Ra \xrightarrow[41\ min]{\beta} {}^{227}Ac$ reaction. The irradiation capacity of the Belgian high-flux reactor, BR-2, is especially appropriate for this type of production, and the required quantities of radium can be made available for a 10- to 15-year production program of 100 to 200 g of ^{227}Ac per year if interest in this isotope warrants. These quantities represent an important fraction of the total amount of long-lived alpha emitters that can be made available in Europe for direct energy conversion. Large-scale production could be started within about 2 years since the necessary production cells can be made available and the studies preparatory to large-scale production have reached an advanced stage.

The principal possible application for ^{227}Ac is as an energy source in combination with advanced thermoelectric and thermionic conversion systems. In a comparison of ^{227}Ac with other alpha emitters — ^{238}Pu and ^{244}Cm, which are usually considered particularly well suited to space applications — ^{227}Ac is found to have a specific power about 25 times that of ^{238}Pu and 5 times that of ^{244}Cm. This high specific power is an important advantage since high-temperature systems with small volume produce the high heat fluxes required for high-efficiency thermionic conversion systems. The 22-year half-life of ^{227}Ac compares well with those of other isotopes considered for space missions. Even if shielding is required, ^{227}Ac will provide the lightest source in the different power ranges.[16a]

The first theoretical and experimental studies, started in early 1963, were made to determine if industrial production of ^{227}Ac would be a sound proposition, both technically and economically. A preliminary program was set up to study conversion rates of ^{226}Ra to ^{227}Ac as a function of neutron flux and time. Calculations showed the flux and usual duration of irradiation cycles in the BR-2 reactor to be favorable for ^{227}Ac; the ^{228}Th produced is considered a by-product. Hardness of the neutron spectrum to which the ^{226}Ra is exposed is important since ^{226}Ra has a resonance in the energy range just above the thermal energy, and thus a favorable contribution to increase the production of ^{227}Ac can be expected from the important epithermal neutron flux of the BR-2 reactor.

Several series of small radium samples have been irradiated with fluxes between 1 and 4×10^{14} n/(cm^2)(sec), most of the samples being placed in the inner space of the BR-2 fuel elements where the epithermal flux is higher. These experiments confirm the substantial increase in the ^{227}Ac production rate contributed by the epithermal neutron flux. The thenoyltrifluoroacetate extraction was used to separate the first irradiated samples, and the various elements — ^{226}Ra, ^{227}Ac, and ^{228}Th — were determined by gamma and alpha spectrometry. Cost estimates from the available production and separation data indicated the cost in dollars per thermal watt to be of the same order of magnitude as those indicated for ^{238}Pu and ^{244}Cm; no value was assigned to the by-product ^{228}Th.

A more detailed program was then initiated to determine production possibilities in the BR-2 reactor and to study the technological problems involved in the irradiation, treatment, and separation of larger quantities of materials. De Troyer described this program in some detail.

A prototype program on production of ^{227}Ac is now in preparation. Approximately 50-g quantities of radium will be irradiated under conditions identical with those required for industrial production. On the

basis of irradiation possibilities of the BR-2 reactor and quantities of radium that could be made available, an annual production of 100 to 200 g of ^{227}Ac can be envisioned. This quantity represents an equivalent annual production of 2.5 to 5 kg of ^{238}Pu, and this level of production could be maintained for 10 to 15 years. About the same quantity of ^{228}Th, expressed as thermal watts, will also be produced concurrently. The production of gram quantities of ^{227}Ac for the first time will make possible the complete study of actinium and its compounds in addition to studies of ^{227}Ac as an energy source.

De Troyer concluded that (1) the results of the prototype program should enable large-scale production of ^{227}Ac if and when required; (2) the cost per thermal watt should be of about the same order of magnitude as that of ^{238}Pu and ^{244}Cm, with ^{227}Ac ranking first because of its higher power density and lower weight per electrical watt; and (3) this annual production gives ^{227}Ac a foremost place among the promising alpha-emitting isotopes that may be required for a possible European space program.

Large-Scale Application of Radiation

FOOD PRESERVATION

Josephson[17] described the aspects of the AEC's food-preservation program in which the primary objectives, as in any food-preservation program, are to extend the useful life of food with minimum reduction of palatability and nutritional factors. There are two theories as to how the radiation acts, both of which help explain the lethal properties of radiation: the "direct hit" theory suggests that the radiation either kills organisms outright or disrupts their reproductive mechanisms, and the "indirect hit" theory suggests that the radiation produces unstable secondary products, notably free radicals and peroxides, which relay the disturbance to other molecules in the cell, enlarging the area and scope of injury.

Before production of irradiated food in the United States can be scaled up to commercial quantities for the broad consumer market, approvals are required from the Food and Drug Administration (FDA) and, for mammal and poultry meats, from the U. S. Department of Agriculture (USDA). These agencies have already approved radiation processing of bacon, wheat and wheat products, and white potatoes.

The AEC's food technology program is under the DID, and aspects of wholesomeness and microbiology are directed by the Division of Biology and Medicine (DBM). The DID has 21 contracts with various universities, private and government laboratories, and industrial groups, three of which involve construction and maintenance of irradiation pilot plants; DBM has contracts with 18 groups engaged in feeding studies and basic research. Josephson highlighted the AEC program and listed the following recent accomplishments:

1. A 3- to 4-day shelf-life extension for papayas subjected to 75 krads in addition to hot-water treatment, and a 10-day increase in shelf life for irradiated mangoes
2. A delay in onset of ripening of bananas with a 10- to 35-krad dose, but subsequent ready ripening with standard ethylene treatment and retention of top quality for 2 to 3 days
3. Tenderization of dry navy beans by a dose of 400 krads and a product more easily cooked
4. Faster drying of irradiated prunes and a more tender product, generally superior to unirradiated fruit
5. An 8- to 15-day extension of shelf life of tomatoes irradiated in the "pink" to "full-ripe" stage

Laboratory research on fish and shellfish is proceeding well, and the program is entering into developmental investigations necessary for successful commercialization of radiation-pasteurized seafoods. Sixteen species of fish and shellfish are acceptable in one form or another as radiation-pasteurized products, and most commercial plastic films, previously approved as food packaging materials, are suitable for packaging radiation-pasteurized fish.

Ten radiation facilities are now, or soon will be, in operation as a result of AEC programs, and others are being planned or are in the construction stage.

After describing the Army food-irradiation program, Josephson concluded that the national food-irradiation program has demonstrated the technical feasibility of radiation preservation of a wide variety of food products. Products for which FDA and USDA approvals have been received or are pending are now ready to go from laboratory-scale to pilot-plant and ultimate commercial production. Large-scale uses of the radioisotopes ^{60}Co and ^{137}Cs and of machine sources of ionizing radiation will be required in the future as this new process takes its place among other methods of food preservation.

In the discussion that followed this paper, Josephson answered an inquiry about a recently published article[17a] on radiation damage to carbohydrates and the secondary effects on carrot cells and the result of this publicity on the relation between FDA and his program. He answered that there has been no effect on FDA. Their response was, "We have reviewed the evidence and reaffirm our position that those clearances that we have given are valid and still stand." Josephson briefly reviewed some of the technical aspects of the study and the problem and described some feeding experiments with carbohydrates. The Army's statement to a Joint Committee of Congress said, "Outside of the adverse publicity, this has had no effect on the Army's program, which is proceeding exactly as we outlined it to you last summer in the hearings by JCAE."

APPLICATIONS OF RADIATION FOR NEW MATERIALS

In the large-scale application of radiation for producing new materials, Harmer[18] said that radiation as a processing tool has developed more rapidly for polymerization than for other chemical processes. One of the early entries into the field was the General Electric Company in the manufacture of polyethylene tapes using accelerator electrons. More recently, other organizations have entered the field with such items as polyethylene insulation, circuit boards, and shrinkable tubing. An estimated $\$20 \times 10^6$ worth of these items were produced in 1964. One of the largest single volumes of processed polyethylene is used in the food-wrapping industry; 1964 sales were estimated at $\$6 \times 10^6$, and continued growth for these products is indicated. All these products were prepared with electron accelerators rather than with radioisotopes. In many cases, which source of radiation is most economical is not clear.

Wood–plastic combinations (WPC) made by impregnating a wood with a monomer and then polymerizing the monomer with radiation may be considered a truly new material. Interest in radiation polymerization originated with the Dow Chemical Co. and the AEC. A number of years ago Dow initiated a program in which monomers were polymerized in the presence of wood. They used solvents to swell the wood structure and achieved dimensional stability of the wood.[18a,18b] The AEC work has been with high monomer loadings, which give a dense product of improved physical properties.[18c–18e]

Other new materials may be made by radiation graft copolymerization. Dow Chemical Co. has developed a copolymer of styrene monomer and polyvinyl chloride. The two materials are mixed together and fabricated before being irradiated with gamma rays and then briefly heated. The styrene acts as a temporary plasticizer and aids processing of the otherwise difficult-to-handle polyvinyl chloride. This combination treatment results in the polymerization of the styrene portion as "branches" grafted to a backbone of polyvinyl chloride, thus rendering a stronger material. Dow has also announced that a graft copolymer of polyethylene and acrylic acid is available in pilot-plant quantities.[18f] In contrast to gamma irradiation of the polyvinyl chloride–styrene mixture, the polyethylene is first irradiated with accelerator electrons prior to the reaction with acrylic acid.

One area of polymerization that should not be overlooked is the polymerization of a relatively large number of compounds to a polymeric solid from a crystalline solid under the influence of radiation without passage through a mobile liquid state. Polymerization of many of these crystalline materials follows rather definite orientation lines, and a pack of fibers, oriented along the crystal axis, may be found in certain systems. The first system to display this tendency was trioxane,[18g] and more recently a sulfur analog of trioxane has been reported, which is highly oriented with respect to the original crystals.[18h] These products may certainly be classified as new materials.

Although commercial application of radiation for other chemical processes has been slower than that for polymerization, Dow has initiated the use of a radioisotope, ^{60}Co, as a catalyst in the manufacture of ethyl bromide from ethylene and hydrogen bromide. Their plant capacity is about 1×10^6 lb/year and operations have been very satisfactory.[18i,18j] Esso Petroleum Company, Limited, of Great Britain is commercializing a process for production of a biodegradable detergent using radiation as a processing tool.[18k] Other radiation processes in the pilot-plant stage have been reported.[18k–18m]

At present the readily available sources of large amounts of high-energy radiation include ^{60}Co, electron accelerators which are available off the shelf, and, more recently, high-energy beta-radiation sources. In the near future, large machines, either electron or X ray, of multikilowatt levels will become available. Cesium-137 should also be available as megacurie sources when the demand develops. For the more distant future, the possibility of using extremely large amounts of radiation directly from nuclear reactors at the site should not be overlooked. Technical problems still have to be solved, but using the direct gamma radiation produced in the reactor core or even the recoiling fission fragments themselves should be possible.

With the coming availability of larger radiation sources, two considerations become apparent: (1) the cost of radiation will probably drop with larger radiation devices, and (2) the capital investment for each level of these radiation devices will probably become larger. The use of radiation in chemical processing and the development of better sources must necessarily grow together. One reason that more radiation processes are not now used in large-scale applications is that a technically sound radiation step does not guarantee that an entire process is economically sound or is acceptable to the marketplace. However, radiation processes now in operation should illustrate that, where industry is willing to invest research dollars in radiation processing, it should, in time, receive a commensurate return on that investment.

RADIOISOTOPES FOR STERILIZATION

Artandi[19] (paper presented by A. A. Stonehill) stated that, before any large-scale use of radioisotopes for sterilization can be established, two basic decisions must be made: (1) Is radiation the best method for sterilization? (2) Are isotopes the best

sources for the process? Radiation is a new and expensive method for sterilization, and justifiable reasons should support its use. Being "best" does not necessarily mean being less costly. It means that, after consideration of all factors that influence process dependability, marketability, and consumer acceptance of the products, the overall effect must be the best for the present and future of the company. Machine sources (electron accelerators) of radiation are applicable for sterilization processes, particularly at higher energy levels (6 to 12 Mev). However, in a recent publication[19a] where electron beams and ^{60}Co industrial sterilization processes were compared, ^{60}Co was preferred because of the inherent simplicity of the process, ease of control, lower maintenance and labor cost, and lower overall sterilization cost.

The choice of an isotope has only limited influence on the design of the radiation plant. At present ^{60}Co is preferred over ^{137}Cs because of greater in-use experience with cobalt and its easier handling and encapsulation, higher specific activity, competitive price, lower self-absorption, and more compact source and facility design. The only factor that favors the use of ^{137}Cs is its longer half-life (30 years vs. 5.3 years for ^{60}Co).

Artandi stressed three factors in selection of a radiation plant design: (1) type of operation, (2) efficiency, and (3) reliability. In some instances batch processes are preferred over continuous ones because they are less costly and simpler and may require less labor, even though they are less efficient in the use of radiation. When there is a well-defined large-scale need, the fully automatic continuous plant may be required.

Efficiency in the use of radiation is important, but not at all levels. Below about 100 thousand curies, using a larger source and simpler design may be less expensive. As the amount of isotope used in a plant increases, efficiency becomes the single most important factor that may justify a more complex and expensive mechanical system. The unscheduled downtime of the radiation plant should be no greater than 10%. Product density, dose uniformity, need for low temperatures, and short process times also influence the design of the sterilization plant.

The sterilizing dose is generally established on the basis of the destruction of a high concentration of selected, extremely resistant microorganisms under carefully controlled experimental conditions. To ensure high reliability, a dose far in excess of that needed for sterilization is used; 2.5 Mrads has been used both here and abroad for 10 years with excellent results.

The most reliable sterilization method, depending only on exposure to the required dose, is irradiation with isotope sources. Before the applicability of radiation sterilization can be established, however, radiation effects on each new product must be studied chemically, physically, and biologically.

Most of the large-scale radiation sterilization has been for medical supplies—sutures, disposable plastic syringes, disposable needles, blades, and catheters—and experimental or pilot-plant radiation sterilization is being applied to a variety of other medical products. Goat hair has been radiation sterilized on a production basis in Australia, and sterilization of animal hides, skins, and pelts is being studied.

RADIOGRAPHY

In introducing the subject of radiography, Coleman[20] stated that this application will never generate the megacurie requirements of some of the other applications. Of nearly 14 thousand licenses issued by the AEC, fewer than 800 are for industrial radiography. Source sizes range from less than 1 curie for small firms to quantities totaling over 10 thousand curies for large radiographic installations, and single sources of more than a thousand curies of ^{60}Co are in use. Cobalt-60 and ^{192}Ir are the common radiographic sources.

The Picker X-Ray Corporation has been working with the American Society for Testing and Materials in producing a library of standard radiographs for steel castings covering a steel thickness range of 2 to 12 in. A set of specially made castings with known defects were radiographed with million-volt X-ray machines, betatrons, and ^{60}Co. Radiographs from properly collimated ^{60}Co are as good as, and for some types of casting defects better than, those attainable with the betatron. The principal advantage of ^{60}Co over the betatron is the lower cost, and the principal disadvantage is the longer exposure time required. When higher specific activity ^{60}Co (600 curies/g) becomes available, shorter exposures will be possible.

Thulium-170 has been used successfully in radiographing welds in thin-walled 1-in.-diameter stainless-steel tubing. A source with less than a 1-mm focal spot is inserted in the tube, and the film is wrapped around the weld. Exposures were less than 1 min, and detail was excellent. Because the source was so small, it was safe to handle on the end of a 4-ft insertion and positioning tool.

Although there are many foreseeable uses in radiography for isotopic sources, especially as specific activities become greater and focal spots smaller, radiography will never create the very-large-scale demand for radioisotopes that some of the other applications discussed at this meeting will. In radiography, where a few curies of an isotope could accomplish results not attainable by any other method, isotope cost is of secondary importance.

While users of major quantities of isotopes for process radiation will seek lower costs, the radiographic industry will seek higher specific activity as one of its prime goals.

RADIOTHERAPY

In describing the use of radiotherapy, Bruner[21] indicated that the number of diseases or conditions treated with electromagnetic radiations used to be much larger than it is now. This change is based on the philosophy that unnecessary radiation exposure is to be avoided and that a number of conditions formerly treated by irradiation can now be more effectively treated by chemotherapy or surgery. Radiation should be practiced with every modern scientific aid, but science is still far from displacing the physicians' art. Radiation therapy continues to remain a conservative art because

1. The effectiveness of a particular type of radiation therapy must be assayed from reactions of diseased tissue in the patient. There is a priori uncertainty as to how a tumor will react to radiation, and no two cancers are equally radiation sensitive.
2. No two patients are just alike.
3. A physician has an ethical problem in devising a control series of patients against which a given therapy is evaluated.
4. Cancers in tissues are seldom well-defined nodules; thus, what should be the overall dose to the tumor and how large an area beyond the tumor should be treated?
5. The radiation therapist usually sees the patient after other physicians, surgeons, and chemotherapists have decided they cannot help further. However, since radiation therapy is used as a supplement to surgery, the value of the therapy alone is difficult to assess.
6. One must wait 5 years and preferably longer to decide that the therapy has halted the progress of the cancer, destroyed it, or had any noticeable effects on it.

For these reasons physicians take a conservative attitude toward proposed improvements in radiation therapy.

Deep-therapy X-ray machines with peak energies of 250 to 400 kv and dose rates up to 500 r/min at 1 m are the workhorses of the therapy department if only because they are so standard. Equipment, dose calculations, and operating procedures are about as standardized as possible.

The next most popular radiation-therapy source is the teletherapy machine. Early units used ^{226}Ra, but, after 1950, high-specific-activity ^{60}Co became available and made possible a really functional design. Cobalt-60 gamma rays (1.17 and 1.33 Mev) are superior to machine X rays because cobalt provides a much more precise depth dose with less lateral scatter and skin effects. In addition, the teletherapy machine is simple to operate and remarkably free of mechanical failures. Once calibrated it retains its calibration, and its beam qualities do not change (except for decay). It is the machine of choice for clinics with unreliable power supplies or difficult access to X-ray machine maintenance and repair.

Only two radioisotopes, ^{60}Co and ^{137}Cs, have been used in therapy machines. Of approximately 1600 therapy machines throughout the world that are listed in a 1963 IAEA publication,[21a] only 3% used ^{137}Cs sources. The advantages of cesium are nominal cost, 30-year half-life, softer radiation (which permits a smaller head with easier placement at odd angles and recesses of the body), and a 0.66-Mev gamma ray which provides a satisfactory depth dose distribution and skin-sparing action. The disadvantages of ^{137}Cs are its relatively low specific activity, bulk, and comparatively large beam with a distinct penumbra.

Until recently, deep-therapy X-ray machines surpassed ^{60}Co in the higher dose rates available and the resultant number of patients that could be treated per day. This has been met with ^{60}Co of higher specific activity; the machines have gone from 1 to 3 thousand curies to as much as 8 thousand curies, and one recently is for 11,400 curies. These larger cobalt machines are fully competitive with, or superior to, the 2-Mev X-ray generators required to produce radiations of about the same energy. Extensive isodose charts for ^{60}Co have been published recently.[21b]

Higher energy X rays can be generated with resonance-transformer X-ray machines, Van de Graaff generators, betatrons and synchrotrons, and linear accelerators. There are about 200 such machines in the world, and their 2- to 70-Mev X rays are of interest because depth dose can be more precisely restricted to the tumor and little skin reaction occurs at the entrance and exit ports. Their greatest use is in treating tumors that lie close to critical organs deep in the body. Their high dose outputs permit short patient exposure time.

Higher energy protons, alpha particles, and other particles produced by linear accelerators and cyclotrons have been shown to be clinically useful. An accelerator at Lawrence Radiation Laboratory, Berkeley, provides 910-Mev alpha particles and 340-Mev protons, which the Donner Laboratory has been using to demonstrate the practical utility of this energy range. The pituitary gland has been destroyed with 910-Mev alpha particles, and excellent results have also been obtained by treating this gland and nerve tracks in the spinal cord with ^{90}Y. Bruner also discussed the possibility of using negative pi-mesons, neutrons, and ^{252}Cf in therapy and research.

In the discussion that followed the paper, Bruner answered questions about the extracorporeal blood

irradiator, damage to tissue occurring from alpha or proton irradiation, and the ranges of these particles. With this high-energy radiation, the amount of debilitation in successfully treated patients is surprisingly low.

Applications of Isotopic Power

AEC ISOTOPIC POWER APPLICATIONS

Carpenter[22] defined isotopic power as including all applications in which the decay energy of the isotope is transformed into thermal energy, which in turn is used as a power source. Carpenter discussed space, terrestrial, and marine uses of the SNAP systems, which are electricity-producing power systems; radioisotope thrusters for space vehicle propulsion and attitude control; and a variety of purely thermal applications of isotopes. The AEC's power-systems work has expanded significantly over the past few years, and the FY 1967 budget request for this work is $20.6 million. The AEC is now applying knowledge gained from a first generation of prototype or demonstration systems to a second generation of SNAP power systems (Table 2).

Just as the power levels for space applications are expected to rise over the years, the trend is also evident in terrestrial applications.* Table 3 shows

*See also "International Study of Terrestrial Applications of Radioisotopic Power Sources" by J. E. Carden, page 172 in this issue.

Table 2 SECOND GENERATION SNAP ISOTOPE POWER SYSTEMS

Unit	Application	Power, watts	Weight, lb	Design life, years	Fuel	Status
SNAP-19B	Nimbus-B weather satellite (NASA)	30	30	5	^{238}Pu	Electrically heated generators on test at AEC and NASA contractors; flight scheduled for late 1967
SNAP-21	Deep-sea uses (Various)	10 (10 to 60)	500	5	^{90}Sr	Completed design and component testing in 1965; proceed with system fabrication and test of 10-watt unit; designs under way for higher power
SNAP-23	Terrestrial uses (Various)	60 (25 to 200)	900	5	^{90}Sr	Design and component development under way for 60-watt unit
SNAP-25	Satellite uses	75	35	5	^{238}Pu	Canceled
SNAP-27	Apollo lunar surface experiment packages (NASA)	56	36	5	^{238}Pu	Component development and test begun August 1965; electrically heated generators go on test in fall of 1966; first fueled unit delivery scheduled for April 1967
SNAP-29	Various satellite and lunar probe missions (DOD and NASA)	400 to 500	400 to 500	0.25	^{210}Po	Competitive design studies completed; hardware development phase being initiated
-	Various spacecraft uses	250	-	5	^{238}Pu, ^{90}Sr	Three design studies being performed; controlled intact reentry and ground handling included
-	Thermionic module development	100	-	-	^{210}Po, ^{244}Cm	Phase I design study being performed
-	Various terrestrial uses	0.1 to 1.0	-	2 to 5	^{238}Pu, ^{90}Sr, ^{147}Pm	Complete design studies by June 1966; initiate development by fall 1966
-	Heart pacemaker	1 to 200 × 10^{-6}	<1	10	^{238}Pu	Proposals being evaluated to initiate development by June 1966

Table 3 TERRESTRIAL ISOTOPIC POWER APPLICATIONS

Power level	Isotope selected	Areas of application	Application criterions
Microwatts	^{238}Pu	Cardiac pacemaker, biomedical stimulation	Extended life, reliability, safety
100 to 1000 mw	^{238}Pu, ^{90}Sr, ^{147}Pm	Oceanographic instrumentation, cable boosters, underwater weapons	Costs, ease of handling, reliability, safety
10 to 200 watts	^{90}Sr	Weather stations, buoys, microwave repeater stations, oceanographic seismographs and instrumentation	Costs, extended life, reliability, safety
1 to 10 kw	^{60}Co, ^{90}Sr, ^{144}Ce	Antarctic support stations, deep-sea mining and exploration, military communications	Costs, reliability, safety

four general areas of application and power level. The higher powered systems will develop with the technologies of heat-source fabrication and power-conversion units.

What is the direction of the AEC efforts to provide isotopic-power-system technologies? In addition to problems of producing isotopic fuels, much work must yet be done with fuel encapsulation in anticipation of higher power, higher temperature systems. In many respects thermal insulation materials currently available have limited application; therefore the AEC has initiated a broad development program on thermal insulations to meet the needs of isotopic systems. Reliable heat rejection for isotopic power systems necessitates the development of static pumps, heat pipes, stable coatings of high emissivity, etc. Larger power systems introduce the need for new approaches to thermoelectric or dynamic power-conversion units. The AEC is also sponsoring study on the organic Rankine system, and NASA has been studying the use of an isotopic Brayton system as a power supply. The prime reason for considering dynamic conversion systems is the advantage in efficiency over static systems and the resultant lowering of the isotope inventory with related reductions in the cost, safety problems, thermal problems, etc. The development of very reliable dynamic power systems involves large and costly test programs. The cost for developing the rotating machinery of the dynamic system is greater than that for developing thermoelectric modules. Carpenter believes that this difference in development cost, if applied to the installation of increased isotope production capabilities, would more than offset the unit-cost advantage of the dynamic system's heat source. The net result would be a versatile modular approach to systems at various power levels and an isotopic fuel availability and cost advantage to users of all isotopic power systems. The AEC is cautiously pursuing the dynamic systems for use with isotopes and is concurrently sponsoring various thermoelectric technology programs.

The AEC is cooperating with the Air Force and NASA in the development of radioisotope thrusters using an isotope to heat a propellant. Ground thruster tests are planned to explore thermal performance and operational problems.

Extremely reliable heat sources, which are insensitive to operating environments (except for heat rejection) and independent of external power sources, have many uses. In developing this type of application, AEC has two main objectives: to provide large, 2 to 30 hp, sources of heat energy to propel undersea craft of all kinds and to provide thermal sources for life-support systems in space, oceanographic, and Earth polar-region applications.

Carpenter concluded that very-large-scale production and processing of the most desirable power-producing isotopes are needed, and the AEC's program is to promote and fulfill this anticipated growth of power-systems development.

PROJECTED RADIOISOTOPE USE BY NASA

English[23] reported that a thermal power level of 50 kw and the period of the 1970's had been selected for investigation of problems connected with the design of large isotopic heat sources. Various candidate isotopes were compared on the basis of hazard and weight requirement.

NASA has yet no approved missions that require large amounts of isotopes; however, without some early planning for this future need, the results would be either a lack of the best isotopes when the demand materializes or an expensive crash program of isotope production. Even when the isotopes are available, the extent of their use will depend on the merits of the isotope power systems and those of competitive systems. Chief among the problems of the isotope systems is the hazard associated with the isotopes themselves. Because several isotope sources may be flown, intact reentry rather than burnup in space has been selected as the preferred design criterion.

In the design of isotopic power systems, several problems must be considered. With alpha-emitting isotopes a void volume must be provided for the helium that accumulates, and, in general, void volume exceeds the volume required for the isotope. Void volumes greater than 0.1 cm^3/watt result in a significant weight penalty.

Another problem is the means of rejecting passive heat to compensate for a variety of conditions associated with launching, space flight, and reentry. Cooling by thermal radiation has been postulated to avoid the risk of failure of active cooling methods. Thermal radiation will also function in space in the event of failure or shutdown of the power-conversion system. English continued the discussion with an assumed geometry of a slab with area sufficient to reject all generated heat from a single face. Relations of power density, slab mass, temperature, slab area, and shield weight were discussed.

During temporary loss of radiant cooling, such as during atmospheric reentry or launch-pad fire, heat sinks of materials with high heats of fusion could absorb the generated heat.

The weights of heat source and shield were determined for a specified geometry for ^{210}Po, ^{147}Pm, ^{238}Pu, ^{244}Cm, and ^{90}Sr. For short missions, i.e., approximately 90 days, ^{210}Po has the lowest total weight (source plus shielding). For longer missions at a given power level, the increased amount of ^{210}Po source material and associated shielding increase the total weight to more than for some other

isotopic sources. Among the longer lived isotopes, ^{147}Pm, ^{238}Pu, and ^{244}Cm are competitive.

Availability is a factor affecting the use of isotopes for space missions, and NASA has requested the AEC to produce ^{238}Pu to support one or two missions a year, with a total production of 500 kw of ^{238}Pu by 1980. To extend the valuable supply of ^{238}Pu, NASA will reuse heat sources on the lunar surface, recover them on reentry, and transfer them from one spacecraft to another.

English compared the general characteristics of solar cell and isotopic Brayton power systems operating in a 300-mile orbit about the earth. Isotopic power systems probably do not have any advantage on the basis of cost alone but offer flexibility in mission planning because of independence of the sun and space environment. Large and oriented surfaces need not extend from the spacecraft, and atmospheric drag and interferences with rendezvous maneuvers, communications, and observations are reduced. An isotope system can readily tolerate rotation for creation of an artificial gravity. The goal in designing a competitive isotopic power system is to realize this potential operational freedom within the limits imposed by nuclear safety.

AEROSPACE NUCLEAR SAFETY

Blake,[24] in discussing aerospace nuclear safety, stated that nuclear energy will certainly prove to be indispensable in space; however, safety considerations have probably been the greatest deterrent to extensive use of nuclear power in space thus far. Safety does not just happen—it must be carefully thought out and positive steps taken to achieve it.

Blake then presented a simplified version of an overall aerospace nuclear safety problem in the form of a multiple-path array. For this relatively simple problem, he pointed out there are 27 different paths through this array which must be considered and for which a safety index should be evaluated. For an actual system the number of paths will be more than those represented by this simple array, and some systems can have thousands of separate paths. The probability of exposure is never zero along any path; therefore evaluation will always be necessary.

When critical paths have been isolated, attention can be given to them to reduce the potential hazard by reducing the number of people involved, the severity of the exposure, or the probability of occurrence (the three items in the safety index).

The primary approach to the safe use of isotopic power in space has been the selection of orbital altitudes that have long lifetimes relative to the half-life of the isotope. In a launch attempt there is some probability that a short orbit or an abort will occur. The designer must consider these and other factors and is limited only by his own ingenuity in designing safer systems.

In some instances the situation may arise where a switch in design approach is necessary. A careful analysis may show that an intact design involving a given fuel form is safer than burnup, or the reverse may be true. In making such a switch, the designer is confronted with the problem of comparing the acceptability of a low probability of exposing a small number of people to a high-level hazard with that of a higher probability of exposing a large number of people to a low-level hazard.

SAFETY OF TERRESTRIAL SOURCES

In a paper on the safety of isotopic power sources for terrestrial applications, Aikens[25] emphasized that a realistic radiological safety evaluation of any facility, system, or device requires the evaluator to be well acquainted with its design, operation, and operational environment.

Present economics indicate that isotopic power sources are primarily advantageous in remote locations, and one might hastily conclude that environmental considerations can be neglected, at least for the present. This, of course, is not true since these devices are tested in and transported through areas of relatively high population. For several years there has been considerable work by industry, the AEC, the Interstate Commerce Commission, and other agencies in establishing safety criterions applicable to the transportation of radioactive material. Recently the AEC published proposed Part 71 of Title 10, *Code of Federal Regulations*,* which sets forth design and performance criterions that must be met by a package containing a large radioactive source.† Both normal and test conditions and performance requirements of the packaging under these conditions are listed in the paper.

When the actual environment to which the device will be exposed is known, the necessity of design or administrative safeguards can be decided from the following considerations:

1. Can failure occur within a certain time, necessitating recovery of the device before it could become a public hazard?
2. If the device cannot be retrieved or if its recovery is questionable, is the design satisfactory to ensure that radioactive material will not be dispersed at a rate injurious to the public?
3. Do the design and location of use warrant the use of a surveillance system that will indicate unauthorized tampering with the device?

*Went into effect Aug. 21, 1966.

†A large source is defined as more than 20 curies of ^{238}Pu or ^{90}Sr; 200 curies of ^{144}Ce, ^{60}Co, ^{137}Cs, or ^{147}Pm; or 5000 curies of material in special form.

Aikens' brief summary was that from the safety viewpoint the least hazardous type and form of radioactive heat source consistent with design and use requirements should be used, transport requirements of such devices should be considered during the initial design phase, and design safeguards and adequate administrative controls should be incorporated to ensure that when the device is in its environment of use it will not present a hazard to the general public, under either normal conditions or reasonably foreseeable accident conditions.

In the discussion that followed this last paper of the meeting, two subjects were considered. The first concerned changing, from a designer's point of view, the criterions based on the maximum permissible concentrations (MPC) for isotopes involved, to terms of maximum permissible thermal watts available. The other concerned safety controls of isotopic power sources, which do not seem to fit within the existing regulations. Experience is so limited that such regulation would have to be exceedingly restrictive.

As knowledge is gained safety criterions will undoubtedly develop in terms of regulations necessary for isotopic power programs. One suggestion was that perhaps a specific device for an envelope of missions might be licensed and a similar type of criterions established for transportation regulations.

(R. E. Greene)

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Section

VI

Isotopes and Radiation Technology

Isotopic Power Development

International Study of Terrestrial Applications of Radioisotopic Power Sources

Abstract: The existing and some potential terrestrial applications of radioisotopic power sources are discussed in the draft report of an international Study Group sponsored by the International Atomic Energy Agency and convened in Vienna, Austria, July 11–15, 1966. The Study Group emphasized the applications and health and legal aspects of terrestrial radioisotopic power sources and considered conversion systems, radioisotope selection, and alternative energy sources.

Isotopic power sources* have been, and continue to be, a provocative subject. There are numerous publications pertinent to radioisotopic power sources for both terrestrial and extraterrestrial applications. Researches with thermoelectric generators* for remote-area terrestrial applications, statements of the official philosophy and scope of the U. S. Atomic Energy Commission (AEC) SNAP (Systems for Nuclear Auxiliary Power) program, and U. S. space and terrestrial isotopic generators for selected studies in FY 1967 have been reviewed previously in *Isotopes and Radiation Technology*.[1]

The radioisotopic power sources are finding many fruitful areas of use. Terrestrial applications, in particular, were the topic of concern of a Study Group (see appendix) sponsored by the International Atomic Energy Agency (IAEA), Vienna, Austria, July 11–15, 1966. In this issue of *Isotopes and Radiation Technology*, we are presenting a summary of the draft report of the information presented before the group in the working papers on the possible terrestrial applications of isotopic power sources and on the health and legal aspects of their use in widely varied environments. The review is preceded by discussion of systems for the conversion of the radioisotopic power to electricity, selection of radioisotopes, and alternative energy sources.

*The term "source" refers to the radioisotopic fuel, and the term "generator" refers to the source plus conversion system.

Radioisotopic Energy-Conversion Systems

Long life and minimum maintenance have been the major incentives for the development of radioisotopic power sources for terrestrial uses as well as for the U. S. space program. The power provided by suitable radioisotopic sources can be as high as several watts per cubic centimeter of source material with a useful life of tens of years. The kinetic energy of the electromagnetic or charged-particle products of radioisotope decay is transformed into electricity either directly, or indirectly through a preliminary conversion to heat.

DIRECT ENERGY-TO-ELECTRICITY CONVERSION

The efficiency and reliability of direct conversion systems have not yet been established, and applications are limited to those requiring sources of very low electric power [<1 Mw(e)]. Three possible direct conversion methods include:

1. Collection of charged particles in a retarding field.
2. Generation of ion pairs in the neighborhood of a solid-state $P-N$ junction or in a gas placed between plates of two dissimilar metals (Ohmart cell).
3. Conversion of fluorescent light, resulting from the absorption of primary radiation by a phosphor, in a photovoltaic cell.

STATIC HEAT-TO-ELECTRICITY CONVERSION

Absorption of the emitted particles or electromagnetic radiations by the radioisotope and its container converts the decay energy into heat energy. When this energy is conveyed through a heat engine, such as a thermoelectric or thermionic converter, electricity is produced. These energy-to-electricity conversion systems are also used with other energy sources, such as nuclear reactors. To date, most work with radioisotopic power generators for terres-

trial applications has been in the 0.1- to 100-watt(e) power range, and static heat-to-electricity conversion systems satisfy these power requirements. Since only a relatively small percentage of the heat generated by the radioisotope becomes electricity, conversion efficiencies (Table 1) are of considerable importance.

Table 1 CONVERSION EFFICIENCIES AND POWER LEVELS OF RADIOISOTOPIC ENERGY-CONVERSION SYSTEMS

Conversion system	Power level, watt(e)	Overall generator conversion efficiency,* %
Direct energy to electricity		
Secondary radiations	<0.001	<1
Static heat to electricity		
Thermoelectric		
Low temperature	<0.0001 to 100	3 to 5†,‡
Intermediate temperature	0.1 to 100	3 to 5†,‡
High temperature	0.1 to 100	?
Thermionic	0.1 to 100	11 to 13
Dynamic heat to electricity	>500	20 to 40

*The overall generator conversion efficiency is lower than the unit conversion efficiency because of additional heat losses and voltage inversion losses.
†Cascading of the thermoelectric materials is possible, and overall generator efficiencies of up to 10% may be achieved.
‡For power levels greater than a few watts.

Thermoelectric Conversion. Operating on the same principle as thermocouples, a thermoelectric converter (part *a* of Fig. 1) illustrates the Seebeck effect, in which two dissimilar metals that are joined at two junctions held at different temperatures produce a current when the circuit is completed.[2] Part *b* of Fig. 1 illustrates how *P* and *N* type semiconductor materials are connected thermally in parallel and electrically in series to produce a current, *I*, through an external load. Thermoelectric conversion provides the efficiency and reliability required at power levels up to 1 kw. The quantity Q_{in} represents the heat flow entering the thermoelectric device from the heat source at a hot-junction temperature T_h. The waste heat Q_{out} must be rejected, leaving the cold-junction temperature of the couple at T_c. This remaining thermal energy must be dissipated in the environment by a method suitable to that environment. In terrestrial systems, where the heat may be lost through radiation, conduction, or convection, the application dictates the choice of heat sink and the means of energy release.

The voltage output of a single thermocouple on load is typically about 0.05 volt. For efficient inversion, outputs of several volts are required, necessitating the use of a large number of thermocouples. For high-power generators [~50 watts(e)] the thermocouples may be mounted separately, but for low-power generators the thermocouples are arranged in clusters or modules to achieve miniaturization.

The semiconductors, such as lead telluride, now being used as thermoelectric materials, offer a significant improvement in efficiency over the simple thermocouple. Thermoelectric materials may be conveniently classified into three temperature ranges:

1. Low, to 250°C, where the bismuth telluride materials have the highest conversion efficiency. Above 250°C the increase in intrinsic conduction resulting from the small energy gap causes increased thermal conductivity and reduced voltage output and leads to a pronounced decrease in conversion efficiency.

2. Intermediate, up to 500 to 600°C, where materials based on lead telluride, germanium telluride, silver–antimony telluride, or lead–tin telluride are used. The upper temperature limit is fixed by the chemical stability of these materials, particularly in air. In an inert atmosphere and with suitable encapsulation, temperatures up to 500°C are possible. (With the lead tellurides there is a small initial falloff in power, which is possibly caused by operating temperature and atmosphere, methods of making contacts, and doping levels. Some manufacturers compensate for this by aging the semiconductors before sale.)

3. High, up to 1000°C, where germanium–silicon alloy and various other silicides are used. These

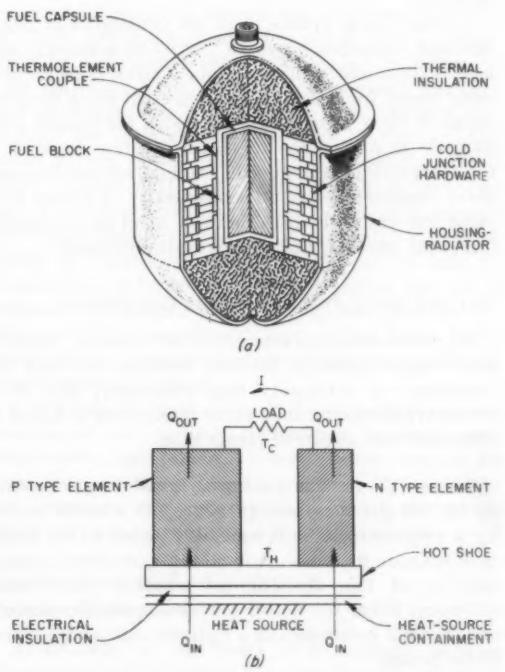


Fig. 1 (a) Cutaway sketch of radioisotope-fueled thermoelectric converter. (b) Schematic illustration of P and N type semiconductor materials connected electrically and thermally to produce a current.

materials are strong mechanically, but problems, such as compatibility and containment of the radioisotopes, are created by the high temperatures.

Thermionic Conversion. Thermionic conversion translates heat into electricity by emission of electrons from the surface of a conductor at high temperature.[2,3] At this surface, heat energy is converted to additional kinetic energy of the electrons within the conductor; increasing the temperature raises the level of this kinetic energy. Electrons that acquire sufficient energy to overcome the forces binding them to the heated conductor (the emitter) escape from the surface. The emitted electrons form a cloud of negative charges that would repel subsequently emitted electrons and force them back to the emitter except for the addition of a gas containing positively charged particles to neutralize the electrons. The mixture of positively and negatively charged particles is the plasma. If the conductor is heated when in proximity to a cooler conductor, the electrons emitted from the heated surface flow to the cooler surface (the collector). Some of the energy of the electrons is imparted to the collector as heat; the remainder provides electron flow (electricity) to an external load circuit. A typical thermionic converter is shown in Fig. 2.

A conversion system with the possibility of highly efficient operation (up to 15 to 20%) at relatively high heat-source and radiator temperatures is potentially very useful for space applications, but there does not seem to be a strong case for using thermionic generators in preference to thermoelectric generators for terrestrial applications. The high source-temperature requirement and the complexity of source construction due to the high thermal-input power density offset the advantage of high overall efficiency.

DYNAMIC HEAT-TO-ELECTRICITY CONVERSION

At power levels greater than 500 watts(e), dynamic heat-engine systems become feasible and have the advantage of extremely high efficiency; this is of considerable importance since it becomes possible to minimize fuel as power levels rise.

Rankine Cycle. In the liquid-metal-to-gas Rankine cycle, the primary energy source is a boiler heated by a radioisotopic heat source. Coupled to the boiler is a dynamic Rankine-cycle power-conversion system (part *a* of Fig. 3). The hot metallic vapor (often mercury) from the boiler drives a miniature turbine and is then condensed in a radiator and pumped back to the boiler.

Brayton Cycle. Whereas the Rankine cycle uses a liquid metal as working fluid, the single-phase Brayton cycle uses a gas, usually argon (part *b* of Fig. 3).

Stirling Cycle. The Stirling (Philips engine) cycle is similar to the Brayton cycle in that a gas is used as the working fluid, but the prime mover in the Stirling cycle is a reciprocating rather than a rotating engine.

A Rankine-cycle prototype and a Stirling-cycle experimental prototype are currently in the design stage as terrestrial radioisotopic power generators in Germany and the Netherlands, respectively. Ex-

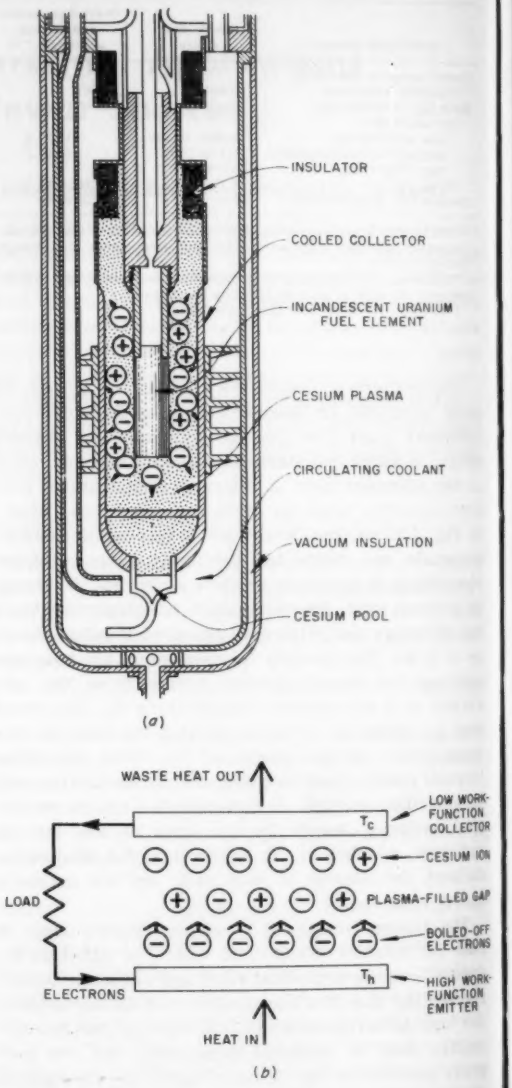


Fig. 2 (a) Typical radioisotope-fueled thermionic unit. (b) Schematic illustration of "boiling-off" of electrons to produce a current through an external load.

tensive Brayton-cycle and Rankine-cycle technology programs for space applications are under way and, along with thermoelectric and thermionic conversion systems, are considered to be the power systems of the greatest potential interest for future U. S. space missions.[4]

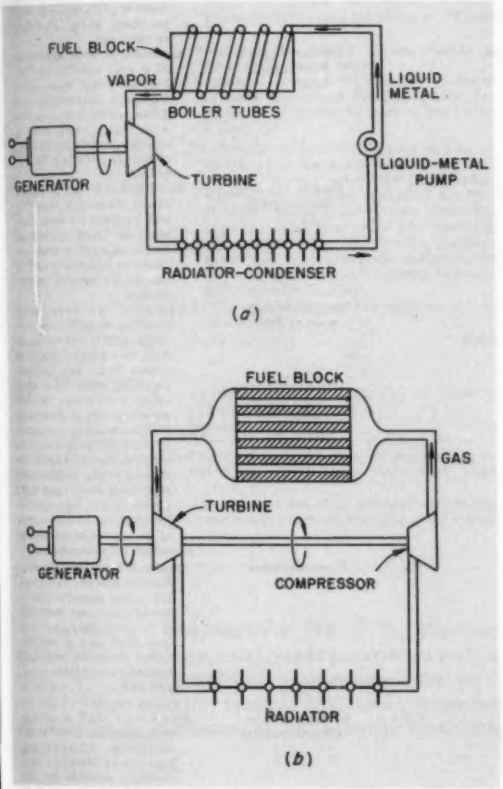


Fig. 3 Two common dynamic heat engines: (a) the Rankine cycle and (b) the Brayton cycle.

Radioisotope Selection

The selection of an energy-conversion system from the above eight will be determined by the particular application and the power-level requirement. The selection of a radioisotope for a terrestrial application will be determined by half-life, specific power and shielding requirements, cost, availability, chemical form, and ease of fabrication. In Table 2 are listed the properties and the advantages and disadvantages relative to ^{90}Sr of a number of radioisotopic sources suitable for power generators. Estimates of the availability and cost of some of these isotopes are given in Table 3.

These tables show that, at present, ^{90}Sr is superior to other isotopes for terrestrial applications in the 0.1- to 100-watt(e) range; ^{90}Sr, ^{60}Co, and mixed fission products (MFP) are preferred for large power sources greater than 500 watts(e); ^{238}Pu is the choice for low-wattage medical applications [<0.001 watt(e)].

Alternative Energy Sources

Radioisotopic power generators are only one type of power-generation system. Alternative energy sources include wind power, solar power, liquid or gaseous fossil fuels, nuclear reactors, and primary batteries.

Wind and solar sources are inexpensive but are not reliable as continuous sources of energy and must be supplemented by some form of storage system, usually batteries. They are also restricted to suitable climatological sites. Fossil fuels, such as diesel oil, propane, and butane, may be used in conjunction with thermoelectric, thermionic, or dynamic converters and with internal combustion engines and fuel cells. Frequent fueling and maintenance and insufficient lifetime and reliability are the primary disadvantages. Nuclear reactors are unlikely to be competitive except at very high power levels [>100 kw(e)]. Primary batteries of either the Leclanché or air-depolarized type are currently in use and are of proven reliability. They are suitable only for power levels up to a few watts and require relatively frequent refueling.

On the other hand, radioisotope-powered thermoelectric conversion systems have demonstrated reliability and maintenance-free lives of greater than 40,000 hr and power levels up to 100 watts(e). Their initial capital cost per electrical watt is high, but their cost on a watt-hour basis compares favorably with other energy sources for applications in remote areas. In relatively accessible regions the cost of transporting alternative energy supplies can be of considerable significance, and in inaccessible regions, such as the poles, desert, or jungle, the additional costs of transporting materials and of servicing increase tremendously.

A full cost analysis shows that radioisotopic power generators are already competitive in price with other power generators for very remote regions. As technology develops and the costs of radioisotopes and complete systems decrease, the instances in which they are competitive will rapidly expand.

Applications of Radioisotopic Power Generators

At present, on the basis of economics, isotopic power sources are primarily advantageous in systems that are difficult to refuel. Applications in remote locations include marine and aircraft navigation aids, meteorological stations, and communications systems; other relatively inaccessible devices are medi-

Table 2 PROPERTIES OF SOME RADIOISOTOPES SUITABLE FOR POWER GENERATORS

Radio-isotope	Half-life, years	Specific power		Compound form	Power density, watts/cm³		Types of radiation	Origin	Comments
		Curies/watt(t)	Watts(t)/g		Theo-retical	Experi-mental			
$^{90}Sr/^{90}Y$	28	150	0.95	$SrTiO_3$	2.35	1.05	β	Fission product; fission yield 5.77%	Available in large quantities at low cost from fission waste currently used in $SrTiO_3$ form, which has a very low solubility in water and brine (an extremely valuable property for safety considerations); later Sr_2TiO_3 and SrO may be used; heavy shielding is required
^{137}Cs	30	207	0.42	Cs glass	-	0.24	β,γ	Fission product; fission yield 6.15%	Available in large quantities at a cost similar to ^{90}Sr, but there are some difficulties in fabrication as a glass; unlikely to compete with ^{90}Sr; power density one-fourth that for $SrTiO_3$; heavy shielding is required
^{147}Pm	2.67	2270	0.37	Pm_2O_3	2.03	1.9	β	Fission product; fission yield 2.6%	Short half-life but a higher power density than ^{90}Sr and requires only one-sixth as much shielding; more expensive and unlikely to compete with ^{90}Sr except for special applications
^{60}Co	5.3	65	17.4	Metal	48	14 at 100 curies/g	β,γ	Reactor irradiation of ^{59}Co	Suitable for very large sources if sufficient activity (>200 curies/g; in fact, Savannah River in Aiken, S. C., has experimentally achieved a specific activity of 700 curies/g) can be achieved; heavy shielding required in inverse proportion to the specific activity; if the source has sufficiently high activity and total power output, less shielding than by a ^{90}Sr source of the same thermal power output may be required
Mixed fission products	Not defined	-	-	Oxides or glass	-	0.1	β,γ	Fission product	Requires aging to give predictable power characteristics; low specific power; inexpensive and available in large quantities due to increased use of nuclear power reactors; chemical and safety problems exist; possible use for large power sources
^{227}Ac	21.2	-	15.0	Ac_2O_3	145	104	α,β,γ	Reactor irradiation of radium	Requires some shielding; unsuitable for medical applications; expensive; high power density; particularly suitable for use with thermionic converters; does not appear likely to compete with ^{90}Sr
^{238}Pu	89	30	0.56	PuO_2	5.6	3.9	α	Reactor irradiation of ^{237}Np	Minimal shielding required; high cost but suitable for low-power generators, particularly for medical use; may require high purity for the latter application
^{232}U	74	26	4.8	UO_2	46	33	α,γ	Reactor irradiation of ^{232}Th	Hard-gamma radiation; expensive; good power density; can operate at high temperature, thus suitable for thermionic converters; unlikely to compete with ^{90}Sr
^{228}Th	1.91	24	170	ThO_2	1530	1270	α,γ	Reactor irradiation of ^{227}Ac or more likely combined with ^{232}U as decay product of latter	Short half-life; requires shielding; expensive; unsuitable for medical applications; very high power density; unlikely to compete with ^{90}Sr
^{244}Cm	18	30	2.8	Cm_2O_3	27	26.4	α,n	Reactor irradiation of ^{243}Pu	Some shielding required so unsuitable for medical applications; expensive; good power density; unlikely to compete with ^{90}Sr

Table 3 ESTIMATED PRODUCTION AND COST OF SELECTED RADIOISOTOPES*

Radioisotope	Country	Estimated production† and cost‡
^{90}Sr and ^{137}Cs	U. S.	100 Mc available; production capability of Isochem, Inc.,§ Richland, Wash., 30 Mc/year [200 kw(t)/year]; similar capability for ^{137}Cs; \$0.50/curie for 0 to 5000 curies ^{90}Sr and \$0.50/curie for 0 to 10,000 curies ^{137}Cs [see *Isotop. Radiat. Technol.*, 3(1): 79 (Fall 1965) for further information on price schedules for larger quantities of ^{90}Sr and ^{137}Cs]
	U. K.	10 Mc/year could give prices similar to those in the United States
	France	Proposed 1 Mc/year ^{137}Cs in 1969 followed by some quantity of ^{90}Sr later; costs expected to be similar to those in the United States
^{60}Co	U. S.	\$0.40/curie at specific activity of 1 to 15 curies/g [see *Isotop. Radiat. Technol.*, 3(2): 173-174 (Winter 1965-1966) for further information on price schedules for ^{60}Co at higher specific activities]; production facilities exist to produce >200 curies/g, but the material is not currently available even to U. S. commercial concerns; the material may become available, but costs are difficult to predict
	U. K.	\$0.70/curie at specific activity of 5 to 10 curies/g
Mixed fission products	Europe	\$1 to \$5/watt(t)
^{239}Pu	U. S.	75 kc/year [2.5 kw(t)/year] at \$900 to \$1800/watt(t) by 1970–1971
	Europe	Similar to United States

*The values presented are included in the draft report prepared by the International Study Group and do not, in all cases, reflect the latest prices and availabilities.

†Minimum production figures based on actual or proposed processing plants; ^{90}Sr in fission wastes from power reactors will total considerably more.

‡No end-of-life value included.

§See page 199, this issue (Miscellaneous).

cal pacemakers and controls (Table 4). Potential applications of radioisotopic power generators include power sources for watches; transponders for temperature, gravity, etc.; seismic devices for detection and warning of earthquakes; and cathodic protection devices.

MARINE NAVIGATION AIDS

The majority of marine navigation lights are in the 1- to 10-watt range and are situated in sites not readily accessible. For example, Sweden today has approximately 1000 small acetylene lights, many of which are located far out in the archipelago; maintenance involves long voyages and high costs. Transport of the heavy acetylene cylinders for recharging involves large ships whose operating costs often exceed the cost of the acetylene in the cylinder. In an investigation by the Swedish Board of Shipping and Navigation, primary contenders as alternative energy sources are (1) primary batteries, which at \$0.04/amp-hr are competitive with acetylene when transport costs are considered; (2) fossil-fueled thermoelectric generators, which are in the prototype stage; and (3) radioisotopes.

The first step in the investigation of the operational capabilities of radioisotopic power generators is the installation in the autumn of 1966 of the 1-watt Ripple-6 (*R*adio*i*sotope-*p*owered *p*rolonged-*l*ife *e*quipment) generator as a minor light in the Stockholm archipelago. Manufactured by the U. K. Atomic Energy Authority (UKAEA), the Ripple generators are powered by $^{90}SrTiO_3$. In Ripple-3, the experimental prototype for Ripple-6, the isotope is doubly encapsulated in cylindrical Hastelloy cans. Energy conversion is by two Bi_2Te_3 thermocouple modules situated at each end of the fuel container. The cylindrical surfaces are surrounded by thermal insulation, and the whole generator is shielded by a tungsten alloy. The Ripple-6 generator is shielded by depleted uranium. Another scheduled 1966 Ripple-generator installation is that of the 1-watt Ripple-5 on the British coast.

A thermoelectric SNAP generator (SNAP-7A), with an initial power of 10 watts and a design life of 10 years, has been in operation since December 1961 (Ref. 5) in a U. S. Coast Guard flashing-light navigational buoy near Curtis Bay, Md. Although the generator still has sufficient energy to flash the buoy light, this is the only SNAP generator that has experienced steady power degradation—to 5.5 watts since 1963—and the cause of the gradual decline in power is of particular concern.

SNAP-7B, a 60-watt 4600-lb thermoelectric generator, has successfully completed a 2-year test at Baltimore Lighthouse in Chesapeake Bay, Md. As the sole source of power to the light, the generator has demonstrated the feasibility of a completely automatic, unattended lighthouse. This same 225-kc ^{90}Sr-powered generator has been relocated to replace the first commercially used radioisotope generator (SNAP-7F) on an off-shore oil platform in the Gulf of Mexico.

Cost estimates for ^{90}Sr-powered generators in the United Kingdom range from \$3000 per electrical watt for 1 watt(e) to \$1500 per electrical watt for 50 watts(e). In the United States a 25-watt(e) generator is on the market for approximately \$60,000. None of these figures include an end-of-life value for the radioisotope. Both the United Kingdom and the United States anticipate at least a 50% price reduction when quantity production is achieved.

Whereas the SNAP-7A and SNAP-7B generators were designed for use on the surface of the sea, other generators have been designed for use under the sea. Radioisotopic power generators in the 0.25- to 5-watt range are being considered for underwater sonar navigation beacons—for channel marking or for spot locations such as oil-well heads. A 7.5-watt(e) generator (SNAP-7E) was installed in an underwater acoustic beacon 750 miles east of Jacksonville, Fla., and 3 miles deep into the Atlantic Ocean and has operated continuously since July 1964

Table 4 TERRESTRIAL RADIOISOTOPIC POWER GENERATORS*

Designation	Application	Radio-isotope	Initial power, watts(e)	Design life, years	Current status: Initial operating date	Current status: Operating time, hr	Shield
United States							
Sentry	Automatic weather station	^{90}Sr	4.7	2	1961	43,800	-
SNAP-7A	Navigation buoy	^{90}Sr	10	10	1962	39,400	Depleted uranium
SNAP-7B	Fixed navigation light	^{90}Sr	60	10	1963	29,160	Depleted uranium
SNAP-7C	Automatic weather station	^{90}Sr	10	-	1962	41,600	Depleted uranium
SNAP-7D	Floating weather station	^{90}Sr	60	-	1964	35,000	Depleted uranium
SNAP-7E	Undersea sonic beacon	^{90}Sr	7	10	1964	35,000	Cast iron
SNAP-7F	Navigation light on oil platform	^{90}Sr	60	-	1963	14,000	-
SNAP-15A	Activator in control system	^{238}Pu	10^{-3}	4	1965	-	None
SNAP-21	Deep-sea sonic generator	^{90}Sr	10	5	1966	-	-
SNAP-23	Lighthouse	^{90}Sr	60	5	1967	-	-
LCG-25 A,B	Oceanographic measuring station	^{90}Sr	25	5	1966	-	-
United Kingdom							
Ripple-1	Demonstration navigation light	^{90}Sr	75×10^{-3}	-	1965	-	Lead
Ripple-2	Demonstration navigation light	^{90}Sr	75×10^{-3}	-	1965	-	Tungsten alloy
Ripple-3	Experimental generator	^{90}Sr	0.75	-	1966	-	Tungsten alloy
Ripple-4	Submarine cable repeater	^{90}Sr	-	20	Design	-	-
Ripple-5	Fixed navigation light	^{90}Sr	1	5	1966	-	Depleted uranium
Ripple-6	Fixed navigation light	^{90}Sr	1	5	Construction	-	Depleted uranium
Ripple-7	Fixed navigation light	^{90}Sr	1	5	Construction	-	Depleted uranium
Ripple 8 to 12	Various	^{90}Sr	5 to 100	-	-	-	-
Euratom†							
EURIG-2	Demonstration prototype	^{170}Tm	5	-	Construction	-	Lead
France†							
-	Demonstrator	^{210}Po	0.3	-	Construction, 1966	-	-
-	Demonstrator	^{90}Sr	12.5	-	-	-	-
Marguerite	Undersea generator	^{90}Sr	0.04	-	-	-	-
-	-	^{90}Sr	-	-	-	-	-
Germany†							
-	Prototype generator	^{147}Pm or ^{3}H	10^{-6}	-	Construction		-
-	-	-	10 to 100	-	Design	-	-
-	-	-	-	-	Design	-	-
Netherlands							
-	Experimental prototype	^{90}Sr	1000	-	Design	-	-
USSR†,‡							
Beta-I	Automatic weather station	^{144}Ce	5.6	-	1964	-	-
Beta-II	Automatic weather station	^{90}Sr	5 to 7	10	In operation	-	-

*Thermoelectric converters are used in all except the German and Dutch generators. The conversion systems used by the Germans are photovoltaic conversion in the ^{147}Pm- or ^{3}H-powered prototype, thermoelectric and thermionic conversion in the 10- to 100-watt(e) generator, and Rankine (organic) conversion in the remaining generator. In the experimental prototype being designed by the Dutch, a Stirling heat engine is used.

†Euratom, France, Germany, and the USSR are also constructing generators for space applications.

‡Information about radioisotopic power generators presented at the Third International Conference on the Peaceful Uses of Atomic Energy, Geneva, Aug. 31–Sept. 9, 1964. No further information was submitted to the Study Group.

as a navigation aid for surface ships and submarines (Fig. 4). SNAP-7E is very similar to SNAP-7A, with a cast-iron shield that is necessary to withstand the 8-thousand-psi pressure, instead of a depleted uranium shield. Another deep-sea unit (SNAP-25) is under design to withstand pressures up to 10 thousand psi and to yield 10 watts. Further development is to include 20- and 60-watt deep-sea units.

AIRCRAFT NAVIGATION AIDS

In international aircraft navigation the trend is toward self-contained airborne navigation systems, satellites, and buoy-borne beacons and away from ground beacons. When ground beacons are used in international navigation, they are located in accessible positions and have high electric-power requirements, thus eliminating any consideration of radioisotopic power generators. However, areas such as Greenland require navigation beacons in inaccessible locations for domestic flights. In such cases, where unattended operation is necessary, low-power-output generators may well be acceptable and economical.

METEOROLOGICAL STATIONS

Automatic weather stations record meteorological information, such as atmospheric pressure, wind direction and speed, air and sea temperature, cloud height, and rainfall, and transmit the data at fixed

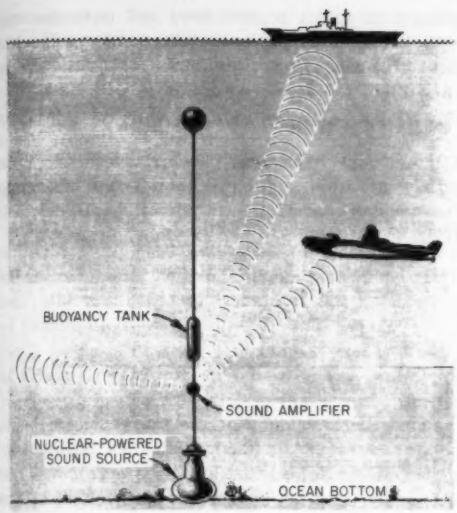
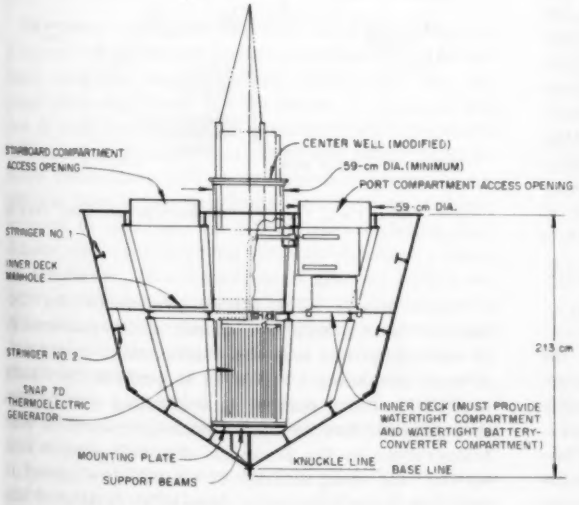


Fig. 4 Schematic of application of SNAP-7E in an underwater acoustic beacon.

times or upon interrogation to manned weather stations. The automatic stations are located in remote areas; some have even been dropped by parachute! Of necessity the equipment must be rugged and capable of withstanding severe climatic conditions. For instance, in the Antarctic where one design of weather station operates efficiently at temperatures down to −54°C, icing, a frozen wind vane, or snow covering the whole station can affect the reliability and usefulness of the measurements. In other regions, dust or salt on the sensing elements will cause instrument inaccuracies, and there are always the general problems of corrosion, variable resistance to strong winds, and, in marine environments, high waves as well.

Power is required for some of the meteorological sensing instruments as well as for 8 to 24 daily radio transmissions that last from less than 1 up to 5 min each, depending on the weather station. The lighting and heating of instruments create additional demands for power. Most power supplies for automatic weather stations are in the 10- to 100-watt range, although a Japanese model used 0.3 watt, and a U. S. model, 5 kw.

In August 1961 the world's first radioisotope-powered automatic weather station began transmitting observations—from Sherwood Head on uninhabited, desolate Axel-Heiberg Island in the Canadian Arctic—to a manned weather station 255 miles to the south-southwest at Resolute Bay, Canadian Northwest Territories.[6,7] Transmissions of observations of wind speed and direction, barometric pressure, and air temperature at 3-hr intervals provided previously unobtainable information. A 5-watt(e) [180 watt(t)] ^{90}Sr-powered thermoelectric generator



(a)

(b)

Fig. 5 (a) Sketch of interior of (b) NOMAD barge type automatic weather station.

(Sentry) continuously supplied the electrical needs of the automatic station established jointly by the U. S. and Canadian weather bureaus. The power from the 17.5-kc strontium titanate source was converted by 60 pairs of lead telluride thermocouples. Physically the weather station consisted of the radioisotopic power generator and the data-telemetry package in an insulated cavity 8 ft below the surface of the ground. (Another advantage of radioisotopic power generators was demonstrated by the use of the 95 watts of waste heat from the generator to maintain a 5 to 27 °C cavity temperature as opposed to the −25 to −1 °C temperature of the permafrost layer.) The wind instruments and thermometer were mounted on a 38-ft tower, and the barometer was positioned in the telemetry package. The power supply functioned satisfactorily for the 2-year design life, but the meteorological sensors experienced several failures. A failure in one of the timing devices in the data-telemetry package resulted in the decision in August 1963 to return the package to Washington for major overhaul and maintenance. The generator will probably be used with modified electronics for further testing at another remote site.

A 10-watt SNAP thermoelectric generator (SNAP-7C) has operated successfully since installation by the U. S. Navy in February 1962 at McMurdo Sound, Antarctica, as the second automatic-weather-station generator. SNAP-7C and SNAP-7A, a marine light buoy, are identical except for the ultimate application and the previously mentioned steady power degradation experienced by SNAP-7A.

Unmanned floating stations have been used by the U. S. Navy for years, but battery replacement has always been a serious problem. SNAP-7D, a thermoelectric generator identical to SNAP-7B, provides 60 watts(e) to a power-conditioning unit, which in turn supplies about 30 watts(e) to a NOMAD (Navy Oceanographic and Meteorological Automatic Device) barge type weather station (Fig. 5) anchored in the Gulf of Mexico.[3] This weather station has operated successfully since installation in January 1964, in spite of two hurricanes, and is a forerunner of a network of such stations planned by the U. S. Navy.

COMMUNICATIONS SYSTEMS

Telephone, telegraph, cable, and radio are the most common long-distance communication systems. Power requirements range from a few watts for cable repeaters to many kilowatts for some transmitters. Communication links often follow devious paths because of the need to place repeaters in accessible and serviceable positions. Underwater cable repeaters are normally spaced at 20-mile intervals along the cable, and power is usually supplied from the end of the cable, thus entailing additional well-insulated conductors. Radioisotopic power generators are being designed to fulfill remote-area and undersea communication-system power requirements in the 1- to 100-watt(e) range.

MEDICAL PACEMAKERS AND CONTROLS

Cardiac and diaphragm pacemakers, heart pumps, auditory and limb-stump prostheses, and bladder and blood-pressure controls are potential medical applications of radioisotopic power generators.

Pacemakers. Demand for pacemakers in both Europe and the United States is currently about 10,000 yearly. The pacemakers now implanted are powered by special mercury cells with a 2- to 3-year design life. A 90- to 400-μw power requirement and the need for a power supply with a longer design life to reduce the frequency of the dangerous implanting operation have led to the design of a radioisotopic power generator for use in a pacemaker. Weighing 97 g, measuring 6 by 5 by 2.8 cm, and yielding 162 μw, the 0.2-g ^{238}Pu-powered thermoelectric generator (Fig. 6) has a 10-year design life plus a 1-year shelf life. Availability of experimental models is scheduled for early 1968, and in vivo tests will then be conducted for 16 to 19 months.

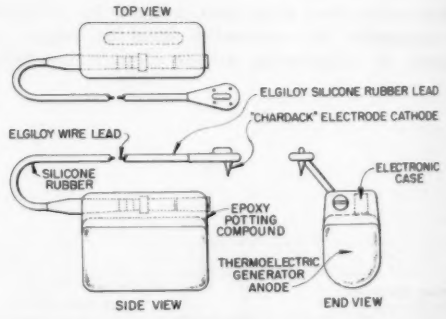


Fig. 6 External view of radioisotope-powered cardiac pacemaker.

Heart Pumps. The strong preference for implanted heart pumps is a result of the unreliability of external power systems. Radioisotopic power generators are being considered to provide the 7 watts needed for heart pumps. In implanting such devices there are severe problems such as control of body temperature in the presence of a concentrated heat source and compatibility of construction materials with the body fluids. No successful devices of this kind have yet been made, either for partially helping the heart or for pumping all the blood supply, and it could be as much as 10 to 15 years before anything of practical value is produced.

Prostheses. The possibility of developing a miniature microwatt radioisotopic power generator to amplify electrical signals in, and to transmit signals from, a prosthetic stump to the motor that drives a prosthetic device, such as a mechanical hand, is being investigated.

MISCELLANEOUS APPLICATIONS

A large number of miscellaneous potential applications are at various stages of development and evaluation.

Watches. Radioisotopic power generators are being developed to meet the requirements of 10 μw of power, a design life greater than 5 years, a price of $25 to $100, and hopefully, a reliability greater than that of current energy sources.

Diving-Suit Heating System. Swim-suit heating has been listed as one of the miscellaneous potential applications of radioisotopic power generators. A data sheet released by a U. S. firm describes the proposed manufacture of a $^{170-171}$Tm-fueled diving-suit heating system in which a thermoelectric generator provides all the electric power required for the system's heat-transfer fluid pump and xenon flash-lamp beacon.[8] The system is designed for 4- to 24-month missions at depths to 600 ft and will sell for between $5000 and $10,000 each, depending on mission duration specified.

Health and Safety Considerations

NONMEDICAL APPLICATIONS

The use of radioisotopic power generators has been governed by safety standards established on an ad hoc basis between manufacturers, users, and the appropriate authorities. But the future development and use of radioisotopic power generators will be greatly influenced by international health and safety implications. Generators will be transported across international boundaries, positioned in a variety of areas, exposed to accidents or severe environmental conditions, and recovered for ultimate disposal; control is mandatory. In essence there are two health and safety controls: (1) integrity of containment and (2) restriction of external radiation exposure. Each proposal for the application of a generator must be carefully assessed in terms of transportation, site, conceivable accidental or environmental conditions, and disposal, so as to ensure adequate implementation of these controls.

Basic rules[9] for the control of radiation exposure and for the international transportation of radioisotopic power generators[10,11] already exist.

Extrapolation of these basic rules provides guidelines for control of health and safety in location, accidental or environmental exposure, and disposal. For example, the transport regulations include detailed standards for design of a package, including physical and mechanical tests that the package must withstand without loss of the radioactive contents. In designing such tests, consideration was given to the accidents in which the package might be involved. Similar standards could be applied to ensure the integrity of the radioisotopic power generator.

MEDICAL APPLICATIONS

Just as for the nonmedical applications, the essential health and safety controls for medical applications are integrity of containment and restriction of external radiation exposure, not only as a consideration for other persons but as a protection for the patient. Special attention must be given to the chemical form and the isotopic purity of the radioisotope. The movement of persons with implanted units constitutes the import and export of radioactive material. Recovery of the unit from a person who expired at home or abroad raises many problems, such as identification of a person with such a unit. For these special problems there are no existing rules.

Recommendations

The Study Group recommended that the IAEA (1) urgently prepare, on an international basis, a preliminary statement of safety principles to ensure an adequate standard of health and safety in the design, installation, use, and disposal of radioisotopic power generators; (2) bring the several problems involved in the medical application of radioisotopic generators to the attention of appropriate international organizations with a view to working out internationally acceptable solutions to the complex problems raised at the meeting; and (3) bring the information contained in the Study Group report to the attention of relevant international, national, and regional bodies with a view to informing them on the likely areas of use of radioisotopic power generators.

(Joan E. Carden)

Appendix

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Section

VII

Isotopes and Radiation Technology

Applications in Life Sciences

Nuclear Medicine—A New Discipline*

By Herbert Vetter,† M.D.

Abstract: "Nuclear medicine" is defined as a special field in which radioisotopes are used for diagnosis. The present status and probable future of this discipline are discussed.

Before discussing the present status and likely future of nuclear medicine in the world, one must first define the term. It seems better not to consider — as some do — nuclear medicine as including the science of radiation effects on man, particularly those likely to result from nuclear warfare. This discipline I would call radiation medicine or human radiobiology, but not nuclear medicine. Nor do I think that the use of isotopes in fundamental medical research is a part of nuclear medicine; the reasons for this separation will appear later. What remains, then, is nuclear medicine as the science of the use of radioisotopes for diagnosis and treatment of disease and in clinical research work.

In this discussion, however, a further narrowing of the subject is required. The uses for cancer treatment of ^{60}Co and ^{137}Cs in teletherapy machines and of ^{198}Au as seeds, ^{90}Y as pellets, and radiotantalum as wires for implantation in tumors are, of course, also therapeutic applications of radioisotopes. In several textbooks and bibliographies, references to this type of work are considered as falling within the field of nuclear medicine. However, there is no doubt that radiotherapy with solid or sealed radioisotopes will remain in the hands of the radiotherapy specialist, where it properly belongs, and it need not be discussed here any further.

Here, then, is a critical point in the discussion. Is nuclear medicine a specialty in its own right? Is there such an animal as a specialist in nuclear medicine? Many people, particularly in England and the United States, deny this vehemently. They argue that radioisotopes are just a tool, like the microscope, and that they should be used by the clinical specialist, e.g., the hematologist and the endocrinologist, along with other tools of his profession. About half a century ago, nearly the same argument was brought forward by those who opposed the establishment of radiology as a specialty. Certainly X rays are also only a tool — in fact, the analogy to the microscope appears to be more appropriate — but their most efficient and most beneficial use soon required a specialist physician who made radiology his main business. Since then this development has continued to a stage where it has become necessary to split radiology even further into radiotherapy and diagnostic radiology.

What are the arguments that speak in favor of nuclear medicine as a specialty in its own right? First of all, the field continues to expand rapidly; new isotopes and labeled compounds as well as new equipment make possible new applications. Ten years ago it could be said that there was no medical specialty in which radioisotopes had not found some application. Consequently the need arose in many hospitals to establish central radioisotope laboratories that would provide services to several hospital departments and clinics. There are, apart from any medical and scientific reasons, a number of economic and administrative considerations that favor this development. Modern counting equipment has become so sophisticated and expensive that only the most affluent hospitals and universities can afford to have, say, an automatic liquid scintillation counter on each floor — and sometimes several on the same floor — often used only occasionally when a particular research project requires it. Centralizing expensive equipment at one place in the hospital and using it fully results in considerable financial savings. Further, the ever-increasing severity of official radiation-protection regulations requires the introduction of increasingly expensive protection measures and, in particular, the establishment of waste-disposal

*A speech presented at the Second National Congress of Nuclear Medicine at Tel Aviv, December 1965. Reprinted by permission from *IAEA Bull.*, 8(2): 7-13 (June 1966). Because of the varied interpretations of the term "nuclear medicine," the editors felt that Dr. Vetter's article would be of interest to medical isotope users.

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facilities, which are most economically set up in one place. The centralization of isotope purchases offers further opportunities for saving money. Finally, central record keeping on patients who have received diagnostic or therapeutic doses of radioactivity is becoming a necessity; instances where a patient still retaining some radioisotope from a previous diagnostic test in one department is given another radioisotope test in another department are becoming more frequent. Thus, in all but the richest hospitals, the tendency is to centralize the radioisotope service; and the person in charge, who devotes full time to running the service, automatically becomes a specialist in nuclear medicine.

Professional Societies

Recognition of nuclear medicine as a separate discipline expresses itself also in other ways. There are now a large number of textbooks devoted to this particular subject. Several journals in the United States, Germany, Japan, and Korea and two in Italy specialize in nuclear medicine. *Excerpta Medica* (Amsterdam, Netherlands) produces monthly a separate volume of abstracts, and the Gmelin Institute (Frankfurt, Federal Republic of Germany), as well as the IAEA, publishes lists of references in this field. Several professional societies of nuclear medicine are now in existence in the United States, Italy, Japan, Germany, and several Latin American countries, with an estimated total membership of more than 3000. They hold annual scientific meetings, which provide opportunities for exchange of scientific information as well as for discussion of problems of professional status and recognition. There are other nuclear medicine meetings, for example, those organized in Oak Ridge, Tenn., and by the IAEA, and the biannual symposiums in Bad Gastein, Austria, which by now have gained considerable reputation not only for their scientific quality but also for their informal atmosphere. Finally, and very significantly, several universities have actually established chairs of nuclear medicine.

There can therefore be little doubt that this development will continue and will ultimately end in the universal recognition of nuclear medicine as a discipline in its own right. Of course, no one will ever dispute the right of individual specialists to use isotopes for their own purposes provided that they are sufficiently trained. This group of "toolists" includes nearly all those who use radioisotopes in fundamental medical research and some clinicians. An extreme case is the accident surgeon who wishes to use one of the new semiautomatic machines to make a rapid assessment of blood volume. He does not have to be a fully fledged specialist in nuclear medicine. The analogy to the dentist who occasionally takes a picture of a tooth granuloma but need not be a full-time radiologist is apparent. The question now arises as to the qualifications we should expect from a specialist in nuclear medicine and what type of training he should receive.

I do not think that he necessarily has to be a physician; there are several examples that demonstrate that a medical physicist can head a hospital isotope laboratory just as well, if not better. Personally, I would prefer to see a physician in charge of a hospital isotope laboratory; he must, however, have a very solid background in physics and mathematics, and in France, for example, most of the leading nuclear medicine specialists have degrees in both medicine and physics. He should also have a full understanding of, at least, the principles of radiobiology and radiation protection and should have spent some time in a chemical pathology department. Most important, however, he should understand the pathology, diagnosis, and therapy of the various disorders for which he may be asked to provide a particular radioisotope service. Otherwise his role will soon be reduced to that of a technician, in the same way that a radiologist without a solid clinical background soon becomes a mere radiographer. It follows that his training will be long—several years, at least—and that training courses on medical applications of radioisotopes of a few weeks' duration are at best only a prelude to long-term and high-level training leading to specialization.

Under these circumstances the fierce discussion that raged years ago (and occasionally does so even now) on whether the nuclear medicine specialist should be a radiologist, an internist, or a clinician of some other specialization is nowadays becoming largely academic. Likewise a newly established group of nuclear medicine specialists should not be incorporated into an existing Society of Radiology or of Internal Medicine but should, from the very beginning, be set up as a separate body.

Physician and Physicist

A physician in charge of a medical isotope service will always be able to speak on equal terms with his colleagues in clinical departments. While the responsibility for patients sent to him for a particular radioisotopic or diagnostic study will always remain with the senders, his voice will be heard and his interpretation of the results of such a study or test will be given proper weight. Once more, the analogy to the relation between the radiologist and his clinical colleagues is obvious. But, however well-trained in physics and mathematics this nuclear medicine specialist may be, he will never be able to provide first-rate radioisotope services without a medical physicist at his side. It is, of course, true that there are a number of routine diagnostic radioisotope tests, particularly those involving sample measurements,

which can be done without the expert advice of a physicist. Beyond the routine tests, however, this is not always the case. A physician in a well-known hospital was about to publish some sensational new results on bone metabolism that he had obtained with ^{47}Ca when he was informed that for the last year or so he had made sample measurements with a single-channel analyzer aligned on the ^{47}Sc peak. His calcium turnover studies were thus, in fact, scandium turnover studies. The need for advice from a physicist is even more apparent in in vivo measurements, and this is true even for such old and established tests as the uptake of radioiodine by the thyroid gland. In the last 3 years, the IAEA has made an extensive survey of techniques of thyroid uptake measurements in about 200 isotope laboratories. The methodology was more satisfactory in laboratories with a medical physicist than in those without one.

With the ever-increasing complexity of techniques and equipment, a medical physicist will in the future be even more needed in a hospital radioisotope laboratory than now. This is not universally accepted, and many hospitals and even teaching departments have just about come around to recognizing the need for a clinical chemist and have established such a post but are far from accepting the idea that a clinical physicist is needed as well. However, even if there should be the required number of established posts available for hospital physicists, there remains the serious problem of finding the physicists themselves. There are only four countries in the world where hospital physics is a profession of sufficient attraction to young physics graduates: the United Kingdom, Canada, the United States, and Sweden. In almost all other countries, physicists are drawn into industry, physics research institutes, or atomic-energy establishments because of better pay and brighter career prospects. The IAEA spends a considerable fraction of its technical assistance funds to alleviate this difficulty.

Another question that arises is of no less importance to nuclear medicine and its future. There is no doubt that the control imposed on the use of radiation and radioisotopes is an outstanding example of the control of a problem before it has become serious. The prevention of radiation hazard to a third party is an accomplishment we should not make light of, but there is now an increasing tendency to extend this control to the first and second parties, namely, the doctor and his patient. The question is: Should we continue to accept this anomalous situation, which has no parallel in any other area of medical practice?

Restrictions on the Doctor

Obviously the use of radioactive isotopes — because of their, at least theoretical, permanent capacity for causing harm — requires special precautions to prevent injury to a third party, i.e., to individuals using them in the course of their duties as well as to those who may become accidentally contaminated, e.g., patients in the same ward or members of the family at home when the radioisotope is administered to an ambulatory patient. Thanks to the efforts of the International Commission on Radiological Protection (ICRP) and various intergovernmental organizations such as IAEA, there are now available sufficiently detailed codes of practice and data on maximum permissible concentrations in air and water to ensure that the nuclear medicine specialist is supplied with information relevant to the protection of both the occupationally exposed radiation worker and the general public. We simply have to follow these rules and regulations to ensure that any third party will not be exposed to risk, and there could not be any objection to the establishment of some sort of control apparatus which sees to it that these rules are observed.

What we are discussing here, however, is the question of the desirability or, perhaps, permissibility of the administration of a certain radioisotope or labeled compound or of a certain quantity of such material by the doctor to his own patient. This question is one that can only, and should only, be answered by the doctor himself. There is surely no difference whatsoever between radioactive compounds and any other compound that the doctor may administer to his patient for diagnostic or therapeutic purposes except for the rather distant relation of radioisotopes to the atomic bomb and fallout from weapons tests with all the associated psychopathological reactions in the minds of the public, including our nonmedical colleagues. Even the property that radioisotopes possess of increasing the natural mutation rate is shared by many nonradioactive drugs, often administered rather indiscriminately.

It is therefore up to the medical profession to set themselves standards of good radioisotope practice and to see to it that those who are guilty of malpractice through neglect or ignorance are called to order. However, has the medical profession sufficient facts available on which to base a sound judgment? The wide variations in opinion throughout the world as to which doses are desirable or permissible indicate that this is not so. There are many countries where standards of good radioisotope practice do not exist; there are some countries where the required decisions are made by a hospital committee and may consequently show considerable variations between hospitals; and, finally, there are a few countries where governmental regulations are so stringent that they hamper orderly progress of nuclear medicine and go so far as to deprive patients of the benefits they may derive from diagnostic radioisotope procedures.

What is being done to provide the medical profession with the necessary data on doses resulting from radioisotope procedures? The data assembled by the ICRP are clearly not applicable since they relate mostly to continuous exposure of radiation workers but give little information on doses resulting from single-shot exposures. The International Commission on Radiological Units and Measurements (ICRU) recognized the urgency of the problem in 1962 and set up two task groups to deal with it. One, on tracer kinetics, has nearly finished its work, which is designed to elaborate the basic framework of generally acceptable definitions, units, and symbols that can be used in internal dose calculations. The second group is concerned with methods of assessing dose and how these should be used in a standard way. The ICRU is likely to stop at this point and to leave it to others to use the recommended methods for calculating doses resulting from typical tracer procedures. One of these others will be the IAEA, which has begun to collate and evaluate the relevant physical and metabolic data, at present widely scattered throughout the literature. It hopes to produce, in due course, dose figures for normal adults, normal children of various ages, and perhaps patients with fairly typical disease conditions.

The IAEA will go no further, however. It will not make recommendations on doses that should be considered permissible or desirable. That judgment is one that only the doctor can, and should, make in the light of the individual circumstances and on the basis of a careful comparison of the benefit his patient might derive from the particular radioisotope procedure with the possible risk from the resulting radiation dose.

Finally, we may just touch on the problem of routine clinical tests. It is understandable that the clinician, particularly the one who uses radioisotopes as a tool only, may wish to have at his disposal some foolproof radioisotope test which can be done with cheap equipment in a minimum time for a maximum number of patients and which gives a clear yes-or-no answer to a diagnostic question. There are at present only very few radioisotope tests that meet these criterions. There are many more that purport to meet them and have been quickly introduced into everyday routine without a clear understanding of their underlying physiopathological mechanisms and the source and magnitude of the associated errors. A classical example is the liver-function test with the radioiodine-labeled dye Rose Bengal, which by now has almost fallen into disrepute. The original technique was crude, several modifications tried to keep the test simple, and still little is known of the fate of Rose Bengal in the human body under both normal physiological and pathological conditions. Observing the very large number of papers that are being published on the clinical results of radioisotope renography and searching in vain for basic information on basic metabolic data, for quantitative methods of expressing them, and for a discussion of sources of error, we sometimes wonder whether this "test" might not one day also be considered unreliable and misleading. Before a new clinical radioisotope test is introduced and its routine use advocated, a very thorough study of its underlying mechanisms should be made in each case, using the most modern techniques and the most recent equipment. Only after this has been done should attempts be made to develop a simpler procedure that would better suit the needs of everyday routine. (MG)

Investigation of Metabolic Processes with Radioisotopes*

By R. A. Leng and C. E. West†

Abstract: The rate of production of certain biochemical compounds used by sheep in oxidative catabolism and the contribution of these compounds to the energy expenditure of the animal under normal and fasting conditions have been estimated by using radioisotope dilution techniques.

Information on the more important overall reactions in the animal body is necessary for the study of animal husbandry under different physiological and nutritional conditions and of metabolic disorders such as ketosis and pregnancy toxemia in sheep. Since organic substances in the animal body are being continually interconverted, the interconversions and their rates cannot be studied by simple chemical determinations of tissue constituents. However, techniques using radioisotope-labeled compounds have been developed[1] with which the overall processes can be

*This is an abbreviated form of an article that appeared in *At. Energy Austral.*, 9(1): 15–18 (1966) and includes work done by E. F. Annison, R. E. Brown, D. B. Lindsay, and R. R. White; printed by permission of the authors.

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investigated and the rates estimated. The method used at the University of New England in Australia is outlined here to show how entry (or turnover) rates of various substances in the body and their contribution to the total carbon dioxide output, and hence to the energy requirement, of sheep can be estimated for use in improving stock raising.

Method

For measuring the substrate entry rate and overall oxidation rate, the substance under study is labeled with radioactive ^{14}C and infused into an animal's bloodstream. The animal may be a sheep prepared with a tracheal cannula some months before the experiment and held in a metabolism cage. An endotracheal tube is placed in its trachea and sealed with an inflatable cuff around the end, forcing the sheep to breathe through the tube. Both jugular veins are catheterized, and the radioisotope-labeled substrate is infused through one jugular catheter while blood samples are taken from the other. The arrangement of the equipment is shown in Fig. 1.

The introduced tracer material mixes with the total amount of the unlabeled substance in the body, mainly through the circulation and mixing of blood and diffusion from the blood into the extracellular and intracellular fluids. Since the radioisotope-labeled substance cannot be distinguished by the body tissues from the nonlabeled compounds, it is constantly removed from the circulation and replaced by the nonlabeled substance produced in the body or absorbed from the alimentary tract. The level of radioactivity is maintained by a constant intravenous infusion of labeled substance (Fig. 2).

Although this is a very simplified outline, and each substance studied in this way provides its own peculiar difficulties, the technique usually adheres closely to that described.

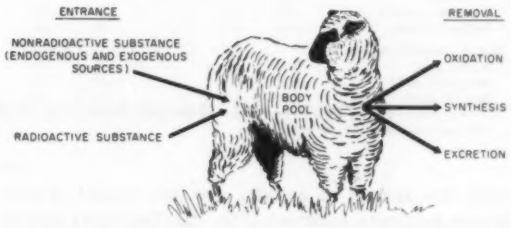

Fig. 2 Entrance of a substance into and its removal from the body pool.

Results

ENTRY RATE OF COMPOUNDS

Entry rates for various blood and rumen constituents in the normal and fasting animal were determined (Table 1). Under steady-state conditions, i.e., when the rate of removal of the substance from the body pool is equal to its rate of entry into the body pool, and after adequate time has been allowed for mixing of the labeled compound with that in the body

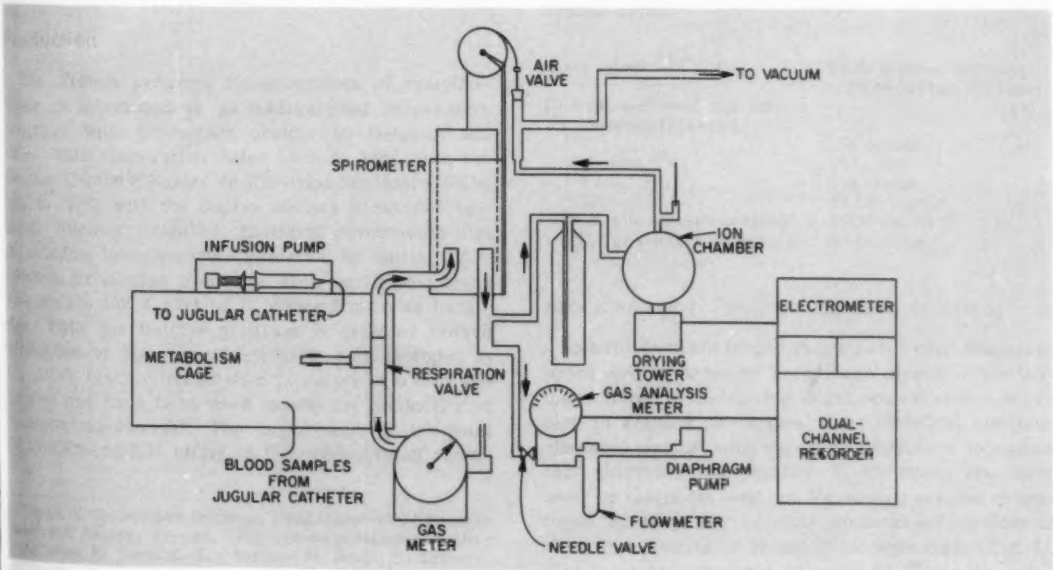

Fig. 1 Arrangement of equipment for studying substrate entry rates and energy expenditures of sheep.

Table 1 METABOLIC DATA OBTAINED ON SHEEP BY RADIOISOTOPE TECHNIQUES

Compound studied	Entry rate, mg/(kg)(min)		Contribution to total CO_2 output, %	
	Fed	Fasted	Fed	Fasted
Blood constituent				
Glucose	2.7	1.5	7	7
Acetate	4.3	1.9	40	20
Propionate	2.75	0.78	*	*
Butyrate	0.60	0	*	*
Formate	0.05	0	*	*
Free fatty acids	0.5	4.2	10	25
Ketone bodies	0.3	0.97	5	10
Rumen constituent				
Acetic acid	6.9	3.4	*	*
Propionic acid	2.3	1.1	21	6
Butyric acid	2.3	1.1	18	4

*Measurements were not made.

pool, the amount of activity per unit weight of substance becomes constant (Fig. 3). The entry rate is obtained by comparing the amount of activity per unit weight of substance over time t with the rate of infusion of radioactivity.

OXIDATION OF SUBSTRATES

When the total carbon dioxide production by an animal is known, the proportion of a substance entering the body pool which is directly oxidized can be calculated. The carbon dioxide content of expired air obtained through the tracheal fistula is determined by passing the air through a katharometer type carbon dioxide analyzer and measuring its radioactivity with an ion chamber and a vibrating-reed electrometer. The total gas flow is measured with a wet gas meter. From these measurements the total carbon dioxide output and the total amount of radioactivity in the carbon dioxide are calculated. Comparison of the amount of radioactivity per unit weight of carbon in the carbon dioxide with that of the substrate in the blood after mixing indicates the contribution of that substrate to the animal's energy expenditure. (MG)

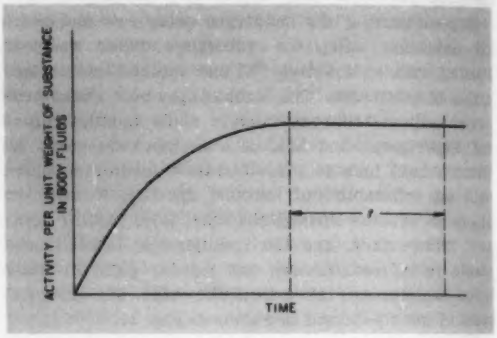

Fig. 3 Achievement of steady-state conditions by labeled compound introduced into bloodstream of sheep.

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Section VIII

Isotopes and Radiation Technology

Isotopes Around the World

Production and Use of Radioisotopes in France

By C. Fisher*

The production and use of radioisotopes in France are managed chiefly by the Commissariat à l'Energie Atomique (CEA). This strong government agency, with minor exceptions, is the only manufacturer in France of radioisotopes, radiochemicals, and radioactive sources. It also maintains a highly competent staff for research and development on industrial uses of radioisotopes and radiation and, through permanent cooperation with private companies, helps to promote utilization of these techniques.

From the beginning, CEA policy has been to consider its activities as a public service, where immediate return for the money invested is not necessarily expected. However, production of radioisotopes has now increased sufficiently that commercial considerations are not only possible but desirable.

Production

The French program for production of radioisotopes is integrated in an international cooperative program with production centers in Belgium and Italy. This cooperation dates back to 1961 with the Belgian Centre d'Études de l'Énergie Nucléaire (CEN) and to 1962 with the Italian Società Richerche Impianti Nucleari (SORIN). Bilateral agreements aim at avoiding unnecessary duplication by setting up a common production program, each participant being responsible for a part of it, depending on its facilities. Thus the Belgian program is oriented toward production of high-specific-activity radioisotopes in the BR-2 reactor; irradiation facilities at SORIN are limited and have been used mostly for production of radiopharmaceuticals. The importance of the joint CEA–CEN–SORIN effort in the production of radioisotopes is indicated by the following figures (as of Dec. 31, 1965):

Number of people employed	200
Number of reactors used for irradiation	9
Laboratory space	6000 m^2
Number of products in catalog	600
Sales in 1965	$1.8 million

The CEA facilities are more extensive and versatile than those of its associates and consequently are adapted to a large variety of productions. The French program includes production of irradiated materials, processed isotopes, radiopharmaceuticals, labeled organic compounds, and sealed sources for industrial uses. For CEA alone in 1965, the following figures apply:

Total sales	$1.32 million, including $0.46 million for export
Quantities of most important isotopes delivered	
^{131}I	459 curies
Colloidal ^{198}Au	152 curies
^{32}P	114 curies
^{192}Ir	13,740 curies
^{60}Co, low-specific-activity	6,100 curies
^{60}Co for teletherapy sources	56,480 rhm

PROCESSED ISOTOPES AND LABELED COMPOUNDS

Quantities much larger than those quoted above are processed in order to keep large stocks of the isotopes listed in the catalog. Radiopharmaceuticals are also in regular production, their chemical and biochemical purity being carefully controlled[1] to ensure high pharmaceutical quality. Single doses are being used in diagnosis with an increasing number of isotopes. In 1965, for example, 20 thousand capsules of ^{131}I with activities of 20 and 50 μc were made (Fig. 1). Sterile plastic syringes of colloidal ^{198}Au with activity from 0.25 to 1 mc are also prepared. The latest

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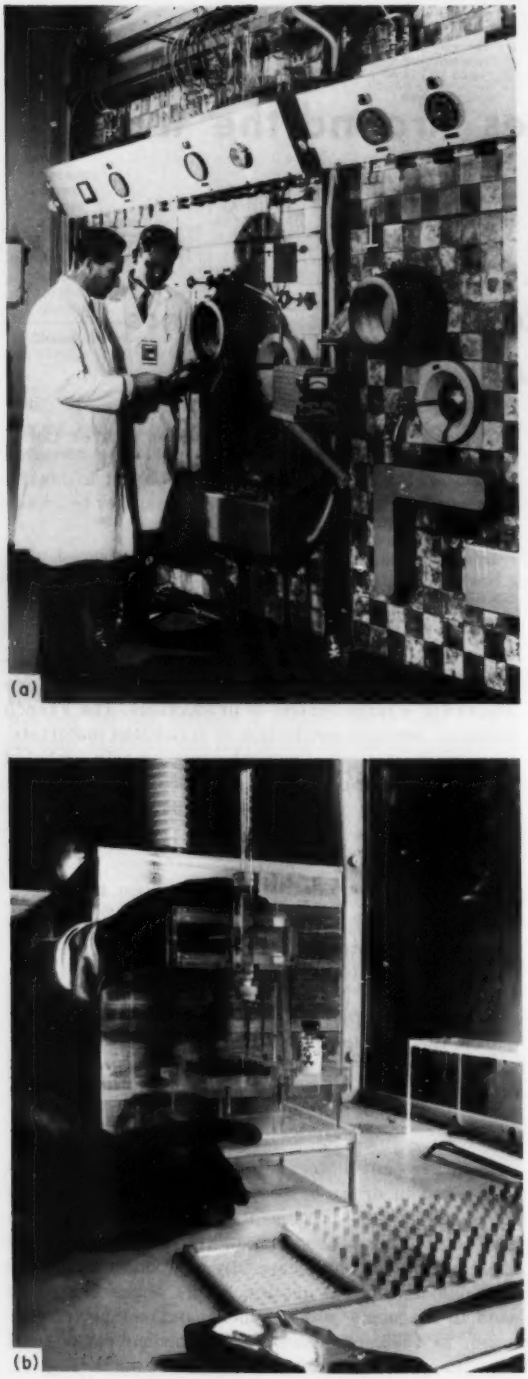

Fig. 1 (a) Production and (b) encapsulation of ^{131}I.

additions to the list of processed isotopes produced are:

^{125}I from natural xenon and from enriched ^{124}Xe
^{28}Mg from ^{26}Mg with a ^{6}Li reaction
^{99m}Tc in multicurie amounts with preparation of ^{99}Mo columns of several hundred millicuries
^{155}Eu from enriched samarium
Colloidal ^{90}Y
^{197}Hg as bromomercurihydroxypropane
^{32}P nucleotides

The 1966 CEA catalog lists 222 labeled compounds, 148 of them labeled with ^{14}C. A 1500-m^2 building for the *service des molecules marquées* has recently been completed at Saclay.

COBALT-60 SOURCES

High-specific-activity ^{60}Co (40 to 100 curies/g) is made by irradiation of 1- by 1-mm pellets or 10- or 20-mm-diameter disks of cobalt in the Belgian reactor. The product is shipped to Saclay, where the sources are assayed, encapsulated, and leak tested.

Low-specific-activity ^{60}Co (10 to 30 curies/g) is obtained from the French experimental and power reactors. Cobalt is irradiated as 50- by 18-mm strips, 1.2 mm thick. The strips are encapsulated in 0.4-mm-thick stainless steel prior to irradiation, and after irradiation they are assembled, as needed, in a second stainless-steel capsule.

An important irradiation program[2] has been set up with Electricité de France, using the EDF-1, EDF-2, and EDF-3 reactors. In EDF-1 and EDF-2 cobalt is used as an absorber and will be removed either when we need it or when it is necessary to compensate for poisoning of the reactor. In EDF-3 the absorber is a part of the fuel assembly and will have to be removed at the same time as the fuel.

FISSION PRODUCTS

The fission-products program has been oriented toward recovery of ^{90}Sr and ^{137}Cs (Fig. 2) in the effluents from the uranium-fuel-processing plants. Separation methods have been studied, principally under a 3-year research contract supported in large part by Euratom.[3] All fission-product solutions available are of the Purex type, i.e., 2*M* in HNO_3, and the process had to be adjusted to use two existing fuel-processing plants, although there may be only one recovery plant. The separation method had to be able to return solutions to the storage tanks without neutralization or reagent addition and with minimum dilution, and operation had to be at the lowest possible cost even for separation on a limited scale. These conditions led to the choice of ion exchange as a separation method.

A mineral ion exchanger—a mixture of 20 wt.% ammonium phosphotungstate with zirconium phos-

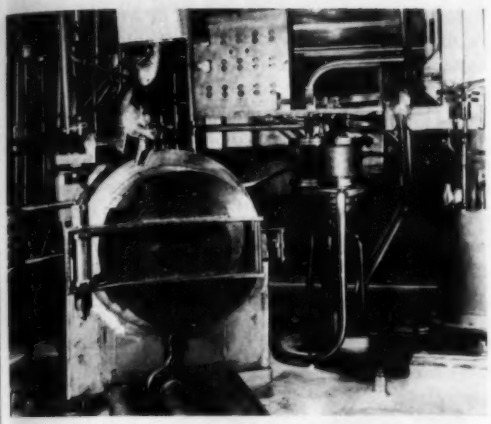

Fig. 2 Pilot unit for ^{137}Cs recovery and purification.

phate—is used for cesium recovery. The ammonium phosphotungstate selectively sorbs cesium in an acid medium, while the zirconium phosphate imparts good mechanical properties to the column. The cesium is eluted with an ammonium salt, which is removed by heating. The cesium that remains is pure enough to be fabricated directly into sources.[4]

Strontium is also recovered with a mineral exchanger—antimonic acid. Preparation of the exchanger still has to be improved for better mechanical stability, but a complete strontium-recovery process is being developed in which the strontium is sorbed directly from the feed solution and eluted by a mixture of nitric acid and silver nitrate.[5]

The production program under development at Marcoule calls for a sorption unit to be built at the fission-products storage plant, where movable columns will be used for sorption of ^{137}Cs or ^{90}Sr in quantities of 100 thousand curies per column. Construction is expected to be completed by the end of 1967. A laboratory for elution, purification, and source fabrication, which will be built at the fuel-processing plant at The Hague, is being designed.[6] With the existing facilities at Saclay, individual sources of up to several kilocuries can be made. Hot cells are under construction for producing strontium titanate sources for isotopic power generators, and a cesium silicoaluminate containing 40% cesium has been developed for use when high resistance to fire and corrosion is required.

Industrial Uses

At the end of June 1966 there were 1959 isotope users in France, 1165 of which were from industry. A survey[7] made in 1962 shows that industrial savings through the use of radioisotopes in 1961 amounted to $8 million. Unfortunately this survey has not been updated, but with the steady increase in the number of users, the figure is now estimated at around $12 million.

The method used by CEA to promote the use of isotopes has been to keep a competent staff and laboratory space available for research and development in cooperation with industry. Money is available for contracts on only a very limited scale ($150,000 in 1966). In the most classical areas of radioisotope use, such as gamma radiography, gaging, and some tracer work, private industry is supplying all the necessary equipment and services. CEA development effort has been concentrated in the following limited areas:

TRACERS AND NONDESTRUCTIVE TESTING

Sediment Tracing: This technique has been developing in France for 12 years, and now several experiments are performed each year in rivers (Fig. 3) and seas by CEA, Electricité de France, and various laboratories. The major recent work undertaken by CEA deals with sediment-labeling techniques and quantitative bed-load-transport measurements.[8] An extensive study[9] is under way on labeling of natural sediments with, primarily, ^{198}Au, ^{51}Cr, and ^{192}Ir. With mud and silt the chief problem is not to disturb excessively the hydrodynamic behavior of the labeled sediments. Several methods have been proposed and are being investigated for quantitative measurement of bed-load transport.[10,11]

Gas Tracing: In a large-scale experiment performed in a Sahara oil field, a hundred curies of ^{85}Kr was injected to trace a gas-lift process. Traces of isotope were recovered 2 years later.

Nondestructive Testing: Several research projects have been undertaken, mainly on gamma but also on beta and alpha radiography. They are concerned with reintensifying screens,[12] image quality and definition, quantitative gammagraphy, and emulsion sensitometry.[13]

Leak Detection: Extensive studies have been made[14,15] on location of leaks in buried pipes, mainly with $CH_3{}^{82}Br$. These studies have been discontinued, and routine leak detections are being made by private companies.

Other Fields: Isotopes are used in wear studies and in continuous measurement of lubricating-oil consumption in internal combustion engines using tritiated oil, which is measured directly by passing exhaust gases into a hot ionization chamber.[16]

RADIOMETRIC ANALYSIS AND CONTROL

X-Ray-Fluorescence Analysis: Light elements are determined by using a windowless proportional counter

containing both an isotopic excitation source (alpha, beta, X, or bremsstrahlung) and sample.[17,18] Peaks corresponding to K beta rays of carbon, fluorine, and oxygen are clearly separated and measured. Heavy elements are determined by using gamma-ray excitation (^{192}Ir, ^{137}Cs, ^{153}Gd) and a scintillation detector.[19] The lead content of ore slurries can be measured in this way. Several studies have also been made on fluorescence excitation by beta rays.[20]

Some problems that have been or are being studied in this group are irradiation technology for routine applications; radiochemical separations[23] using chiefly ion-exchange resins and giving high separation yields and decontamination factors in automatic operation; computer techniques for reducing complex gamma-ray spectrums or decay curves and correcting dead time, with applications to mixtures of short- and long-lived radioisotopes;[24,25] activation analysis applied to high-cross-section matrixes such as rhenium, tungsten, tantalum, and copper; 14- and 3-Mev-neutron applications to instrumental analysis both with and without specific radiochemical separations.[26] This group gives practical training to industrial people and postgraduate students and also accommodates analysts who want to analyze their own samples but do not have their own activation-analysis laboratories.

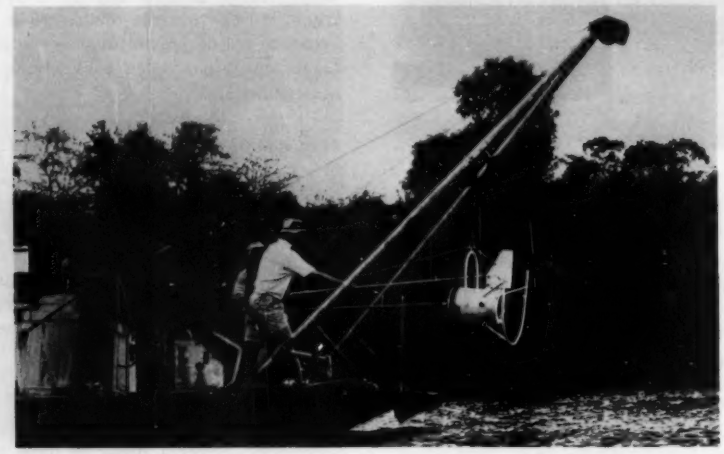

Fig. 3 Positioning of tracer injector in Mekong River (Cambodia) siltation studies.

X-Ray-Fluorescence Coating-Thickness Measurement: An industrial gage has been developed to measure the thickness of zinc coating on steel sheet in a continuous plating line.[21] A laboratory instrument has been developed to measure the thickness of very thin layers of iron–nickel deposited on glass plates for magnetic memories.[22]

ACTIVATION ANALYSIS

The Activation Analysis Group, created in 1959, has the objective of developing activation-analysis techniques and applications to industrial and research problems and providing irradiation and laboratory service to activation-analysis users in France. It has at its disposal the irradiation facilities in two reactors with fluxes up to 2×10^{14} n/(cm^2)(sec), two pneumatic tubes with four irradiation positions between 2×10^{12} and 4×10^{13} n/(cm^2)(sec), one hydraulic tube at 8×10^{13} n/(cm^2)(sec), four gamma-ray multichannel spectrometers, punched-tape readout systems for computer calculations, radiochemical laboratories for low and high activities, and a 400-kv 2-ma accelerator producing high fluxes of 14- or 3-Mev neutrons.

RADIATION TECHNOLOGY

The Radiation Technology Group studies problems that arise in the design, building, and use of industrial irradiation facilities using radioisotopes. It will provide private industry with calculation methods, dosimetry methods, and technological information to enable it to design high-efficiency irradiators.

This group operates a pool-and-cell facility where up to 100 thousand curies of ^{60}Co can be handled. The pool is 5 m long and 2.5 m wide; a canal passes under the cell wall. The cell is 4 by 4 m in inside dimensions. The whole ceiling is removable, and the cell is provided with a window, two master–slave manipulators, and an elevator that goes down to the pool.

The cell will be used for dose measurement in different source configurations, true activity determination, and, eventually, medium-scale irradiations. The pool will be used for storage, irradiation, and infinite-medium dose-distribution measurements. The pool and cell are in a new building providing low-activity experimentation areas, laboratories, and offices.

HYDROLOGY

Important work has been done in river flow-rate measurements using radioisotopes, rates as high as 1000 m^3/sec being measurable. Isotope techniques have been applied to detection of seepage through dams and from canals and to study of pollution problems in seas and rivers. In underground tracing ^{131}I, ^{51}Cr-EDTA, and inactive NaI are used.[27,28]

Because of the extensive use of tritium as a tracer in hydrological studies, we have equipped a laboratory to measure tritium concentrations in water. We have two liquid scintillation counters, one gas proportional counter, and two sets of 10 cells each for electrolytic enrichment.

Hydrological studies are being made in different places by measuring the tritium concentration periodically in rain, river, and spring waters in order to obtain information on the water cycle. Rainwater is also collected at 26 stations in France to determine the geographical tritium distribution in the country as a working base for future hydrological studies. Apparatus for ^{14}C measurement is being set up for dating water from the dissolved-carbonate content.[29]

Three areas are included in apparatus-design research. A depth probe is being tested which can give directly, after standardization and by means of a nomograph system, the moisture, dry density, and chemical nature of the soil. A project is under way to make another depth moisture probe able to give measurements at 5-cm intervals. In the second area, radiography with a neutron generator or a neutron source is being studied with 150- and 400-kv deuterium–tritium generators, a 400-kv deuterium–beryllium generator, and a 3-curie americium–beryllium neutron source. Finally, apparatus based on dilution of cadmium salt solution has been studied for measuring underground-water flow rates. Dilution of the cadmium solution changes the transmission of the radiation from an americium–beryllium source. The rate of an underground current of water in the range 0.5 to 10 m/day can be measured.

COOPERATION WITH INDUSTRY

Cooperation with industry is very effective, the following examples being typical of CEA's policy:

In tracer applications and especially in activation analysis, CEA has concentrated on research and development of new methods. Routine analytical service has been transferred to Société de l'Atome Industriel in laboratories located near the reactors at the Grenoble Research Center.

In X-ray-fluorescence work, problems are studied at Saclay up to the point where a laboratory prototype is operating. Then industrial equipment is developed with private manufacturing companies (Nucleometre, CGEI-Lepaute).

Pilot industrial irradiation sources have been built by private companies, e.g., Saint-Gobain Techniques Nouvelles (SGN) and Conservatome. A mobile ^{137}Cs irradiator (IRMA) loaded with 170 thousand curies is based at Saclay.[30] The Centre Lyonnais d'Applications Atomiques (CLAA) irradiation facility has ^{60}Co in the irradiation cell, put there by CEA, and both organizations use the source. (MG)

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Industrial Use of Radioisotope Devices in Japan*

The reasons given by a number of Japanese industrialists for failure to fully exploit radioisotope devices are summarized here. A survey made in March 1962 on the use of radioisotope equipment in Japan had indicated less than 400 actually in use at that time. In order to find out why the number was so low, a committee of the Japan Radioisotope Association, in January 1964, queried 746 organizations—commercial, educational, and both government and private research institutions (excluding medical)—on performance of this type of apparatus. Of the organizations contacted, 84% replied, and 36% of these were using radioisotope devices. Of the 547 instruments in use, 25% were imported. The percentage of those imported varied widely with the type: 17 and 14%, respectively, of the thickness and level gages, all the vacuum and gas chromatography meters and leak detectors, but none of the snowfall meters. The isotopes concerned were ^{14}C, ^{60}Co, ^{85}Kr, ^{90}Sr, ^{106}Ru, ^{137}Cs, ^{147}Pm, ^{204}Tl, ^{226}Ra, ^{133}Ba, and ^{63}Ni. An abbreviated form of the questionnaire used is shown in Fig. 1. Considerable detail was requested from those who were already using radioisotopic devices.

Organizations Using Radioisotope Devices

In the organizations where these instruments were in use, thickness and level gages each accounted for about 35% of the total number (Table 1). Both were in use with ferrous and nonferrous metals, paper, pulp, and plastics; thickness gages were reported also in rubber, cement, glass, and foodstuff organizations and level gages in use with ceramics and petroleum. The thickness meters were 81% of the beta transmission type, 3% beta reflection, 4% gamma transmission, and 11% gamma reflection. Of the level gages, 44% were used in the plastics industry. This high percentage is considered due in part to the high temperatures used in this industry, which make other measurement methods difficult. More than half the level gages were of the type with source and detector in the same horizontal plane.

Mechanical breakdowns accounted for 20% of the total failures among the thickness and level gages:

Point of breakdown	Thickness gage	Level gage
Mechanical parts (e.g., drive)	13	16
Electrical components	24	29
Electrical cables	7	5
Detectors	2	14
Defective design or manufacture	10	14
Other	1	1

These industrial instruments used to be designed in electrical laboratories, but consultation with mechanical engineers may lead to improved reliability.

Of those who indicated the frequency of breakdowns, a high proportion showed less than five per year:

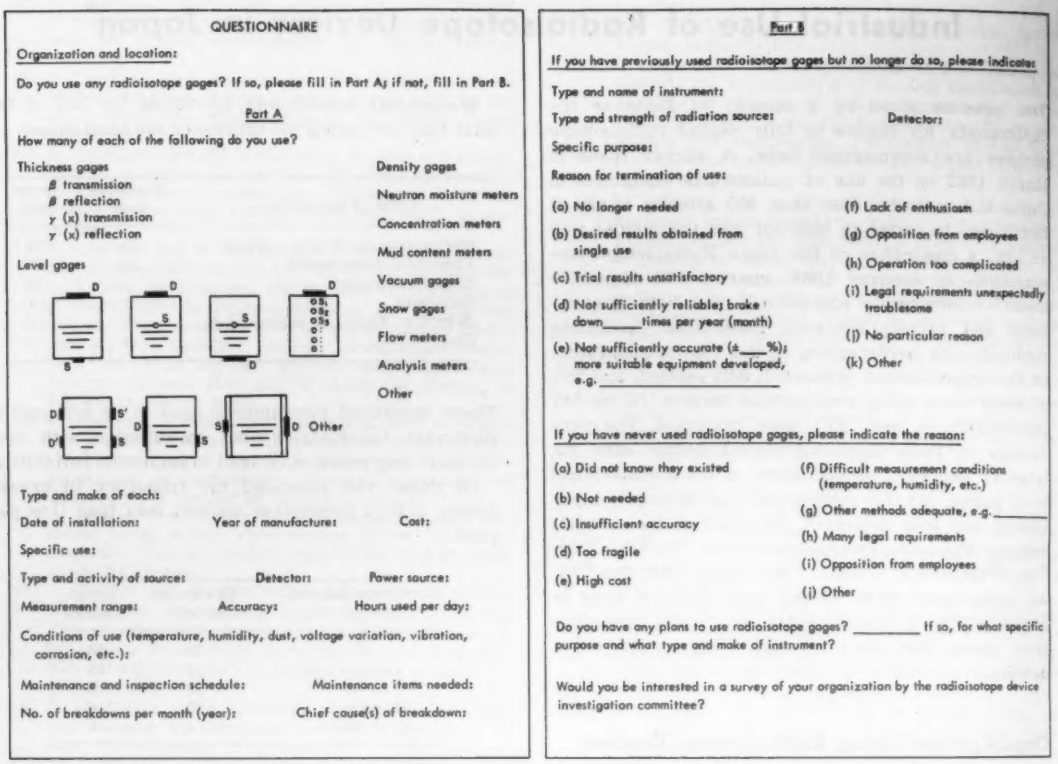
No. of breakdowns per year	Thickness meters	Level meters
0	36	28
1 to 5	91	79
6 to 10	18	9
11 to 20	23	6
21 or more	9	1

These data are interesting, but their usefulness is limited because of the variations both in the amount of time per day the instruments were used and in the conditions under which they were used. For example, the cases of 21 or more breakdowns per year were all for instruments operated continuously, i.e., 24 hr/day. Instruments used in small numbers, like snow gages, broke down very seldom. In some failures reported as breakdowns, the trouble was actually due to electronic-tube aging or battery rundown rather than to any problem in the radioisotope part of the system.

Vacuum meters were the most widely used of the remaining types. They were all based on the variation in alpha-particle ionization with air-pressure change. Steel manufacturers were the chief users, for measuring the vacuum in smelting furnaces, but an electrical equipment manufacturer also reported their use with transformer tanks and mercury rectifiers.

Neutron moisture meters were reported in 24-hr use in the iron manufacturing industry, with two instruments being used for automatic control. Use was intermittent for measurement of soil moisture and other applications. Snow gages were being used principally by electric-power companies. Analysis meters

*Summary of "Report of an Investigation of the Present Utilization of Radioisotope Devices" by T. Naito, I. Otsuka, H. Iijima, H. Imamura, H. Ono, K. Hirooka, H. Fushimi, and Y. Maebashi, *Radioisotopes (Tokyo)*, 14: 255-267 (1965) (English translation AERE Trans-1048 by J. B. Sykes). Earlier Japanese data were included in *Industrial Radioisotope Economics*, Findings of Study Group Meeting, Vienna, 1964, Technical Reports Series, No. 40, International Atomic Energy Agency, Vienna, 1965 (STI/DOC/10/40); summarized in *Isotopes and Radiation Technology*, 3: 152-166 (1966).



QUESTIONNAIRE

Organization and location:

Do you use any radioisotope gages? If so, please fill in Part A; if not, fill in Part B.

Part A

How many of each of the following do you use?

Thickness gages
- β transmission
- β reflection
- γ (x) transmission
- γ (x) reflection

Density gages

Neutron moisture meters

Concentration meters

Mud content meters

Level gages

Vacuum gages

Snow gages

Flow meters

Analysis meters

Other

Type and make of each:

Date of installation: Year of manufacture: Cost:

Specific use:

Type and activity of source: Detector: Power source:

Measurement range: Accuracy: Hours used per day:

Conditions of use (temperature, humidity, dust, voltage variation, vibration, corrosion, etc.):

Maintenance and inspection schedule: Maintenance items needed:

No. of breakdowns per month (year): Chief cause(s) of breakdown:

Part B

If you have previously used radioisotope gages but no longer do so, please indicate:

Type and name of instrument:

Type and strength of radiation source: Detector:

Specific purpose:

Reason for termination of use:

(a) No longer needed
(b) Desired results obtained from single use
(c) Trial results unsatisfactory
(d) Not sufficiently reliable; broke down_____times per year (month)
(e) Not sufficiently accurate (±___%); more suitable equipment developed, e.g._____
(f) Lack of enthusiasm
(g) Opposition from employees
(h) Operation too complicated
(i) Legal requirements unexpectedly troublesome
(j) No particular reason
(k) Other

If you have never used radioisotope gages, please indicate the reason:

(a) Did not know they existed
(b) Not needed
(c) Insufficient accuracy
(d) Too fragile
(e) High cost
(f) Difficult measurement conditions (temperature, humidity, etc.)
(g) Other methods adequate, e.g._____
(h) Many legal requirements
(i) Opposition from employees
(j) Other

Do you have any plans to use radioisotope gages? _____ If so, for what specific purpose and what type and make of instrument?

Would you be interested in a survey of your organization by the radioisotope device investigation committee?

Fig. 1 Abbreviated form of questionnaire used by Japan Radioisotope Association in investigating the use of radioisotope devices.

Table 1 RADIOISOTOPE DEVICES IN INDUSTRIAL USE IN JAPAN IN 1964

Device	Number in use				
	Commercial	Educational institutes	Government research institutes	Private research institutes	Total
Thickness meters	183	2	3	-	188
Level gages	182	1	-	1	184
Vacuum meters	52	4	-	-	56
Density meters, transmission	28	2	3	-	33
Density meters, scattering	6	-	14	2	22
Neutron moisture meters	10	-	6	1	17
Snowfall meters	6	-	4	-	10
Analysis meters	8	-	2	-	10
Leak detectors	7	-	-	-	7
Potentiometers	1	-	4	-	5
Gas chromatograph detectors	5	-	-	-	5
Mandrel safety devices	2	-	-	-	2
Radiation signal devices	2	-	-	-	2
Plug detectors	1	-	-	-	1
Tobacco weight inspection	1	-	-	-	1
Clinker volume meters	1	-	-	-	1
Cosmic ray meters	-	-	1	-	1
Logging equipment	1	-	-	-	1
Air humidity meters	1	-	-	-	1
Total	497	9	37	4	547

were used mainly for determining sulfur and the carbon/hydrogen ratio in fuel oil and petroleum and for analysis of cement raw materials.

Organizations Formerly Using Radioisotope Devices

Some organizations that reported had used radioisotope devices previously but then discarded them. The reasons for discard and the number of times the reason was given were considered among the most significant data obtained and included

Unsatisfactory test results	10
Single use sufficient for desired results	9
No longer needed	6
Troublesome legal requirements	2
Insufficient reliability	2
Complicated operation	1

The types of gages involved here included thickness, level, density, concentration, moisture, vacuum, and snow.

Organizations Never Having Used Radioisotope Devices

It is encouraging that, among those who had never used radioisotope devices, only a few gave employee reluctance and legal restrictions as a reason. Reasons that were given more than once were

Lack of need	184
Adequacy of other methods	50
High cost	17
Insufficient accuracy	11
Troublesome legal requirements	10
Difficult measurement conditions	9
Employee opposition	2
Ignorance of existence of such devices	2

Suggestions for Improving Radioisotope Gages

The use of radioisotope gages in Japan might well increase if certain improvements could be made; a number of them were suggested by the groups who had used or were using such instruments:

	No. of suggestions			
	Thickness	Level	Other	Total
Better performance and reliability	25	6	28	59
Better servicing arrangements	12	6	8	26
Simpler operation	4	-	3	7
Lower price	4	2	1	7
Other	10	7	4	21
Total	55	21	44	120

(Martha Gerrard)

Section

IX

Isotopes and Radiation Technology

Miscellaneous

Conferences

Seventh Annual Meeting of the Southeastern Chapter of the Society of Nuclear Medicine

By H. Hupf*

More than 200 persons participated in the seventh annual meeting of the Southeastern Chapter of the Society of Nuclear Medicine held Nov. 3–5, 1966, in Durham, N. C. C. D. Maynard (Bowman Gray School of Medicine) chaired the scientific program committee, and A. P. Sanders (Duke University Medical Center) served as local arrangements chairman. The meeting was accredited by the North Carolina Academy of General Practice, Commission on Education, as a 17-hr course. Representatives and exhibits from 10 radiopharmaceutical and instrument companies were present.

The renogram was discussed from several viewpoints. C. Harris (ORNL) covered the necessary instrumentation; R. Witcofski and I. Meschan (Bowman Gray School of Medicine) discussed the labeled compounds usually used; and R. Greenlaw (University of Kentucky Medical Center) talked about expected developments. A method of determining the rate of glomerular filtration by external monitoring of ^{51}Cr-labeled inulin was presented by A. Johnson (VA Hospital, Coral Gables, Fla.).

The nucleus of the cell is no longer the only source of interest in radiation damage studies, according to Sanders, who presented evidence of extranuclear cell damage in a lively session on radiobiology in nuclear medicine. D. Pizzarello (Bowman Gray School of Medicine) discussed a radiobiological basis of clinical isotope use; F. Comas (Oak Ridge Institute of Nuclear Studies) talked about the oxygen effect; and D. Zimmerman (University of Florida) presented information on the effect of radiation on rat thymus.

In the session on pulmonary scanning, J. Quinn (Northwestern University Medical School) discussed results of inhalation scanning and related problems and proposed a method of double lung scanning using ^{131}I-tagged albumin by perfusion and ^{99m}Tc albumin by inhalation. D. Sabiston (Duke University Medical Center) talked on perfusion lung scanning and showed movies of emboli formation in animals. In some cases normal pulmonary scans were obtained from animals with established emboli and thrombosis.

Liver scanning with scintillation cameras was discussed by A. Gottschalk (University of Chicago), who also exchanged views with Harris on the relative merits of the camera vs. rectilinear scanner.

The participants expressed a great deal of interest in generator systems that provide radionuclides (half-lives between 1 and 24 hr and gamma emission between 100 and 350 kev) that can be made colloidal or incorporated into biologically active molecules, e.g., ^{99m}Tc, ^{113m}In, and ^{188}Re. Future work will probably be concerned with finding new and better generator systems.

Abstracts of all 57 papers presented at the meeting will be published in the *Journal of Nuclear Medicine*.

AEC Activities

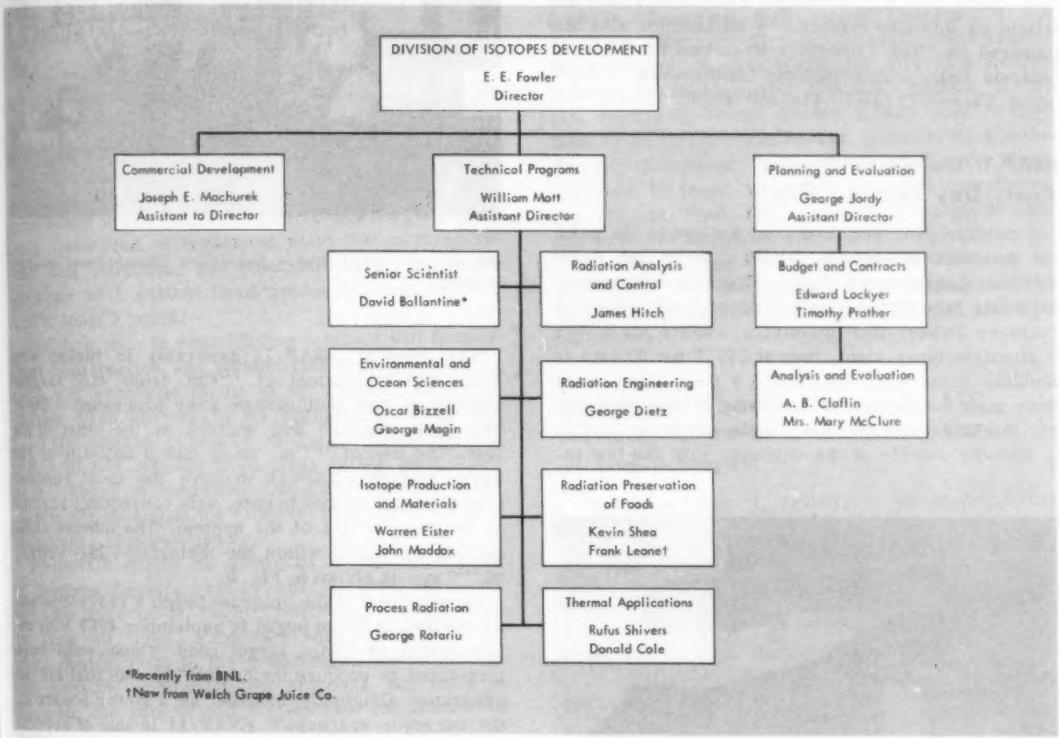
Programmatic Changes in AEC Leave DID Free To Pursue Uses of Radioisotopes

The increasing demand for isotope-fueled electrical generators has required the Atomic Energy Commission to realign program responsibilities for several headquarters divisions. Robert E. Hollingsworth, AEC General Manager, has given new responsibilities to the Division of Reactor Development and Technology (DRD&T) and to the Division of Space Nuclear Systems (SNS) to allow the Division of Isotopes Development (DID) to return to its basic mission.

Conversion of radioisotope heat energy to mechanical energy is now assigned to DRD&T or SNS once the initial development stage is passed. All applications, electrical or mechanical, that relate to the space program will be handled by SNS; applications leading to electrical power systems in terrestrial or oceanographic systems are to be handled by DRD&T. Under the new system, DID will still have full responsibility for research and development directed toward use of radioisotopes as heat sources, when the heat is not subsequently converted to mechanical or electrical energy.

*Isotopes Development Center, Oak Ridge National Laboratory.

DID can now direct its efforts more fully toward increasing the use and variety of other applications of radioisotopes and radiation. The division has also been reorganized (see accompanying chart).



Isochem Asks Termination of FPCE Contract

Isochem, Inc., and the AEC will start negotiations promptly to terminate the contract and land lease under which Isochem was to construct and operate a commercial plant to separate and encapsulate useful fission-product isotopes from reactor wastes at Hanford. At first, the AEC announced an application for delay by Isochem, but a week later the announcement was made that the contract would be mutually dissolved. Isochem based its decision on the lack of a market for its products and rising construction costs for the Fission Products Conversion and Encapsulation (FPCE) plant.

Concurrent with the FPCE construction contract was the contract to operate the chemical separations plant to process plutonium and uranium from spent fuel elements. The AEC is asking for "expressions of interest" from other firms who might wish to operate the chemical processing facilities and would be willing to assist or provide commercial projects for the Richland area. Other bids for the original contract came from Dow Chemical Co. and from United Nuclear Corp. and Monsanto Research Corporation jointly.

ORNL Develops Radioisotope Light Source Photometer

A beta-emitting radioisotope, ^{36}Cl, has been mixed with a scintillating material to provide an unusually stable light source for use in photometric analysis. Because the pulses of light given off by the scintillator are proportional to the isotope's decay rate and beta energy, the spectral and emission characteristics of the system can be very accurately determined. According to the inventor, H. H. Ross of ORNL's Analytical Chemistry Division, there is theoretically no limit to the precision that can be reached with this system.

One of the most desirable features of the system is that the output from the system is digital; this means not only that maximum sensitivity and precision can be attained, but that digital computing equipment can be coupled directly to the analytical instrument. Remote analysis, computer-coupled analysis, space and oceanographic explorations, and trace

determinations of organic material in biochemical processes are all areas of possible application of the technique.

For further information, contact the Office of Industrial Cooperation, Oak Ridge National Laboratory, Post Office Box X, Oak Ridge, Tennessee 37830. An article on this new invention by its inventor also has appeared as "New Concept in Precision Photometric Analysis Using a Radioisotopic Light Source," *Analytical Chemistry*, 38(3): 414-420 (1966).

SNAP-11 Generator Passes Ninety-Day Test at ORNL

A nuclear generator designed for use on the moon has produced electricity for 90 days in Oak Ridge National Laboratory's Lunar Simulation Chamber. Beginning July 12, the SNAP-11 (Systems for Nuclear Auxiliary Power) was operated at −235°F for 5 days to simulate lunar night, then at 235°F for 80 days to simulate lunar day, and finally 5 more days under lunar night conditions. The loading of the generator into the large vacuum test chamber is shown in Fig. 1, and the outside of the chamber with the test instrumentation is shown in Fig. 2. Electrical output was 23 watts at the start of the test under lunar night conditions; during lunar day, the unit produced 18 watts. As the test progressed, the fuel decayed as expected, and the output dropped to 20 watts during lunar night and 15 watts during lunar day.

Fig. 1 Loading the SNAP-11 generator into the Lunar Simulation Chamber.

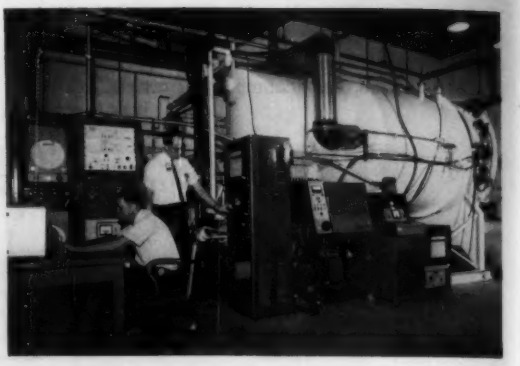


Fig. 2 The ORNL Lunar Simulation Chamber and instrumentation.

The 20-watt SNAP-11 generator is fueled with 7.5 g (25 kilocuries) of ^{242}Cm from the largest amount of this radioisotope ever processed—20 g. The fuel produced 900 watts(t) at the start of the test. The use of ^{242}Cm, which has a half-life of 162 days, enabled SNAP-11 to have the most concentrated power source to date, with consequent savings in the total weight of the system. The intense alpha heating generated within the material—120 watts/g of ^{242}Cm—is shown in Fig. 3.

Development of the program began 4 years ago, and production of ^{242}Cm began in September 1965 with the fabrication of ^{241}Am target rods. These rods were irradiated to produce the curium used as fuel for the generator. Originally intended as a power source on the Surveyor spacecraft, SNAP-11 is one of several in the SNAP series designed for space exploration.

Fig. 3 Two of five ^{242}Cm oxide pellets fabricated to fuel the SNAP-11 generator. Each pellet produces about 200 watts(t).

The generator was developed by the Martin Co. of Baltimore; ORNL produced and encapsulated the fuel and conducted the test.

AEC Asks Industry for Proposals To Build Meat Irradiation Plant

The Atomic Energy Commission, with the Departments of Defense and Commerce, has asked industry to submit proposals on the design, construction, and operation of a pilot plant for meat irradiation. With government help, the facility is to demonstrate the feasibility of commercial production of radiation-processed meat, promote civilian interest, and provide quantities of irradiated meat for military use and test purposes. The plant will have a minimum capacity of 1 million lb of processed meat annually for at least 3 years.

The firm that is awarded this contract will finance plant construction and furnish full reports on technical and economic facets of the project. AEC will provide $140,000 toward the engineering design and $230,000 toward purchase of the radiation source. DOD will buy 300,000 lb of meat during each of the first 3 years of operation. The Department of Commerce will assist in the program to promote civilian acceptance of irradiated food.

Proposals should be submitted to AEC's Division of Isotopes Development by Apr. 7, 1967; the Commission expects to begin preliminary design work by June 30, 1967. Proposals will be evaluated on plant design, cost to the government, experience of the company submitting the proposal, and plans to promote the sale of and test-market the irradiated meat.

Review Panel Appointed by AEC To Review Licensing Procedures

The Atomic Energy Commission has appointed a panel of extra-government experts to evaluate policies and procedures of radioisotope licensing and regulation. The AEC itself wished to simplify its procedures to better serve users' needs without compromising the excellent safety record so far achieved, and it has reached the point where it feels that an outside, independent study would be valuable. The panel will review and appraise policies, procedures, and regulations for licensing and regulating radioisotope use; provide a full report of the panel's views and conclusions as to how effectively the policies and procedures protect public health and safety; and submit recommendations on any improvements that the panel believes are necessary. Recent actions of the AEC improving the radioisotopes licensing program will be given special consideration.

The AEC has appointed the following to the panel:

Mr. George F. Trowbridge, a Washington, D. C., lawyer with the firm of Shaw, Pittman, Potts, Trowbridge & Madden; Dr. Lauchlin M. Currie, Mamaroneck, N. Y.; Dr. John M. Heslep, Department of Public Health, State of California, Berkeley; Dr. Wilfred R. Konneker, Mallinckrodt Chemical Works, St. Louis, Mo.; Dr. Seymour Rothchild, New England Nuclear Corp., Boston, Mass.; Dr. Merrill A. Bender, Roswell Park Memorial Institute, Buffalo, N. Y.; Mr. Donald T. Green, Picker X-Ray Corp., Cleveland, Ohio; Dr. Herbert Parker, Consultant, Battelle–Pacific Northwest Laboratory, Richland, Wash.; Dr. John P. Ryan, Minnesota Mining & Manufacturing Co., St. Paul, Minn.; and Dr. G. Hoyt Whipple, University of Michigan, Ann Arbor.

Mr. Trowbridge will serve as chairman of the panel. Mr. Bruce Churchill of the Isotopes Branch of AEC's Division of Materials Licensing will serve as secretary.

Shalek and Christian Named to AEC Advisory Committee on Medical Uses of Radioisotopes

Chairman Glenn T. Seaborg of the Atomic Energy Commission has appointed Dr. Robert J. Shalek and Dr. John E. Christian as members of the Commission's Advisory Committee on Medical Uses of Isotopes. The Advisory Committee advises the Commission on policies and standards for regulating and licensing medical uses of radioisotopes in humans.

Dr. Shalek is an associate physicist at the M. D. Anderson Hospital and Tumor Institute, University of Texas, Houston; Dr. Christian is Head of the Bionucleonics Department of Purdue University, Lafayette, Ind. The new members replace Dr. Edith Quimby and Dr. Donald Childs who have retired from the Committee. Committee members serve 5 years on a rotating basis under which two members are replaced each year.

Other members of the Advisory Committee are: Dr. Wallace D. Armstrong, Professor, Department of Biochemistry, University of Minnesota, Minneapolis; Dr. Reynold F. Brown, Member, Department of Radiology, University of California Medical School, San Francisco; Dr. John A. D. Cooper, Dean of Sciences, Northwestern University Medical School, Chicago, Ill.; Dr. Robert H. Greenlaw, Associate Professor of Radiology, University of Kentucky, Lexington; Dr. E. Richard King, Professor of Radiology, Medical College of Virginia, Richmond; Dr. George V. LeRoy, Medical Director, Metropolitan Hospital, Detroit, Mich.; Dr. Rulon W. Rawson, Attending Physician and Chairman, Department of Medicine, Memorial Hospital, New York City; and Dr. Harald H. Rossi, Professor of Radiology, Col-

lege of Physicians and Surgeons, Columbia University, New York City.

Nebraska and Washington Become AEC Regulatory Agreement States

The Atomic Energy Commission has approved agreements under which Nebraska and Washington will assume part of AEC's regulatory authority over the use of radioisotopes in these states. Nebraska's agreement was signed by Governor Frank Morrison and Commissioner James T. Ramey in Lincoln on Aug. 29, 1966. The Washington agreement was signed by Governor Daniel J. Evans and Commissioner Wilfrid E. Johnson at Richland on Dec. 6, 1966.

The Nebraska agreement became effective Oct. 1, 1966. The Division of Radiological Health will be responsible for licensing, rule making, and enforcement; the Department of Health will conduct the radiation-control program. There are 105 AEC licenses in Nebraska.

In Washington the Department of Health will be the state agency responsible for licensing, rule making, enforcement, and radiation control. The 190 AEC licenses came under state control on Dec. 31, 1966.

Other states with agreements with AEC are Alabama, Arkansas, California, Florida, Kansas, Kentucky, Mississippi, New Hampshire, New York, North Carolina, Oregon, Tennessee, and Texas. An agreement with Louisiana is pending.

Medical Licensing for Diagnostic Procedures Simplified Under New AEC Proposal

The Atomic Energy Commission is proposing to amend the regulations for licensing the medical use of radioisotopes in diagnostic procedures. Granting licenses to a qualified applicant for more than one related procedure (even if only one is applied for) will greatly simplify the procedure both for applicants and the Commission. The amendment would divide the most common diagnostic uses of radioisotopes into two groups and would specify that an application for one use in a group may be considered as an application for all uses in a group. The applicant must satisfy present AEC medical licensing requirements and must have adequate training and equipment to handle all the uses in the group.

Medical uptake, dilution, and excretion studies make up one group. This includes tests of thyroid, liver, and kidney functions; fat absorption studies; and determinations of volumes of certain body substances. Scanning procedures and tests for localizing tumors are in the other group.

The Commission believes that the new procedure would eliminate hundreds of applications for changes in medical licenses in the first year of operation.

Use of Tritium in Glow Lamps Exempted from License by AEC

A petition from the General Electric Company of Cleveland, Ohio, has resulted in the Atomic Energy Commission's amending its regulations to exempt from license the possession and use of up to 10 μc of tritium in glow lamps. Tritium makes glow lamps start more quickly in areas of reduced light or of darkness. Glow lamps are used in many items ranging from very complicated electronic circuits to simple general appliances. The manufacture or import of glow lamps containing tritium will, of course, still require a license from the AEC.

Availability of Isotopes and Services

ORNL Stable-Isotope Inventory Augmented

Recent additions to the ORNL enriched stable-isotope inventory include the following:

Isotope	Purity, %	Amount available, g
^{26}Mg	99.8	4
^{33}S	48.5	1
^{34}S	67.9	8
^{40}Ca	99.96	40
^{70}Ge	84.6	50
^{72}Ge	90.9	100
^{74}Ge	94.5	100
^{98}Mo	98.7	200
^{114}Cd	98.6	40
^{150}Nd	96.0	4
^{174}Hf	6.7	1
^{176}Hf	77.5	1
^{180}Hf	98.2	11
^{203}Tl	92.3	200

General Atomic Offers Local Activation-Analysis Training

Practical courses that include laboratory demonstrations, lectures, and technical films are now available from General Atomic. These courses are designed to be given at local industrial plants and laboratories. The lectures are presented by V. P. Guinn, Technical Director of General Atomic's Activation Analysis Program. Fees start at $1500 and

are based only on the length of the course, not the number of participants. For further information, contact R. M. Watkins, General Atomic, Activation Analysis Service, P. O. Box 608, San Diego, California 92112.

General

First Commercial Food Irradiator Bankrupted by Crop Failure

The worst potato crop in Canada in 35 years has put the world's first commercial food irradiator, operated by Newfield Products Ltd. at Mont St. Hilaire near Quebec, into bankruptcy. The potato crop was harvested late because of excessive rain, and some potatoes were chilled by frost. Neither waterlogged nor chilled potatoes respond well to radiation treatment.

The loss is put at $500,000. The Bank of Montreal, Industrial Acceptance Corporation of Canada, and other creditors are currently discussing what to do with the facility. Atomic Energy of Canada Ltd. is expected to reclaim the ^{60}Co source (approximately 30,000 curies) leased to Newfield Products and use it in a pilot demonstration.

The facility had irradiated 7 million lb of potatoes, according to John Masefield, vice president of the company, and the product was well received in the market.

Isotope-Powered Water Recovery System Passes Air Force Test with Flying Colors

A radioisotope-powered water recovery system designed for use aboard space vehicles has recently passed a 30-day test. About 500 g of ^{238}Pu was used as a direct heat source in the experimental system that changes human fluid wastes (wash water, urine, and condensate) to potable water. The vacuum distillation–vapor pyrolysis system demonstrated both the direct use of radioisotopes and the dependability of the device when exposed to a space environment for a lengthy period.

Under actual flight conditions the device would recover water over a period as long as 1 year; the half-life of the radioisotope is 89 years. The demonstration device was designed for a 30-day mission during which time it would supply two men with about 0.75 lb of water each hour. The use of radioisotopic heat allowed a simple design, eliminating the need for such things as electrical resistance heaters, lead-ins, and associated controls.

The recovery process starts with distillation of the waste liquid in a vacuum at 150°F; this requires 210 watts(t). The water vapor is then heated to 1200 to 1600°F in the presence of ruthenium to eliminate ammonia, bacteria, and organics; this uses 70 watts(t). Condensation of the vapor provides high-quality drinking water.

The two-man water recovery system is about 30 by 22 by 30 in., and hot or cold water is dispensed in 1- to 6-oz quantities by pushing buttons on the console.

The device was developed for the Aerospace Medical Research Laboratories at Wright-Patterson Air Force Base, Ohio, by General Electric's Missile and Space Division. The test was run at Mound Laboratory, which is operated by Monsanto Research Corporation for the AEC.

Harwell Ships 100 Thousand Curies of ^{60}Co to Turkish Grain Irradiator

The Atomic Energy Research Establishment at Harwell, England, has recently sent 100 thousand curies of ^{60}Co to Iskenderun, Turkey. The large grain irradiator there is designed to hold 170 thousand curies and is a prototype plant that can process 50 tons/hr of maize, rye, or barley. The disinfestation plant is owned by the Turkish firm of Toprak Mahsulleri Ofisio, although its financing is through the Turkish government and the U. N. Development Program. Nuclear Chemical Plant Ltd. of England designed and built the plant under administration of the International Atomic Energy Agency.

(FEM and RHL)

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